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### This is the author's manuscript

*Original Citation:*

*Availability:*

This version is available <http://hdl.handle.net/2318/1566633> since 2019-01-07T11:21:44Z

*Publisher:*

Geological Society of London

*Published version:*

DOI:10.1144/SP410.3

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# UNIVERSITÀ DEGLI STUDI DI TORINO

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In Zellmer, G. F., Edmonds, M. & Straub, S. M. (eds) The Role of Volatiles in the Genesis, Evolution and Eruption of Arc Magmas. Geological Society, London, Special Publications, 410, 143-177, First published online August 6, 2014

By

**Cigolini C., Laiolo M., Coppola D. (2014)**

*The definitive version is available at:*  
*La versione definitiva è disponibile alla URL:*

**<http://dx.doi.org/10.1144/SP410.3>**

# Revisiting the last major eruptions at Stromboli volcano: inferences on the role of volatiles during magma storage and decompression

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## Abstract

Stromboli is a unique open conduit volcano and a natural laboratory to investigate how volatiles migrate and concentrate under dynamic conditions. Fluid phases are involved in magma decompression and pressurization, modulate Strombolian activity and rule magma rise and fragmentation processes. Here, we have revisited the available data on the last two major eruptions at Stromboli volcano and concentrated our analysis on the 2007 eruption. First, we further analysed petrological-geochemical data to assess equilibrium conditions by using standard thermobarometry; then we used a grid of selected reactions which involve solid-melt-fluid equilibria to better constrain the P-T regimes that adequately describe our system. Primitive hydrous basaltic melts, reported in literature, and preserved as melt inclusions in olivine (with 2.3-3.8 wt% of H<sub>2</sub>O, and 890-1590 p.p.m. CO<sub>2</sub>), are equilibrium with forsteritic olivine and a diopsidic clinopyroxene, at average pressures of 260 (±47) MPa for temperatures approaching 1170 (±17) °C, and calculated  $X_{CO_2}^m$  (mole fraction of CO<sub>2</sub> within the melt) ranging from 0.60 to 0.76. Ca-rich or ultracalcic melts are regarded as a result of decompression along a steep adiabatic and/or isothermal curve. During this process the magma will crosscut the stability field of diopside and enter the liquidus field. The earlier crystallized diopside is destabilized and will react with the coexisting liquid phase leading to

30 the formation of ultracalcic melts. Ejected golden pumices (with 2-3 wt% H<sub>2</sub>O), are in equilibrium  
31 with Ca-pyroxene, forsteritic olivine and anorthitic plagioclase at 150–220 MPa and temperatures  
32 of 1120-1150°C. Evolved melt inclusions (substantially degassed) in Fe-richer olivine (ca. Fo<sub>70</sub>) of  
33 the scorias show average equilibration pressures of 78 ( $\pm$  20) MPa, and temperatures 1138 ( $\pm$  14)  
34 °C.

35 In summary, the higher *P-T* regimes associated with the origin of primitive melt inclusions are  
36 representative of the base of the chamber, where the ferromagnesian phases may crystallise and  
37 cumulate. The magma with a bulk composition typical of the pumices is stored the middle and main  
38 part of the chamber (likely its axial sector) and these materials are erupted during paroxysmal, and  
39 more rarely, major explosions. Finally more evolved melts inclusions found in the olivine of the  
40 scorias are indicative of crystallisation within the conduit or its root-zone connected with the upper  
41 part of the chamber.

42 Pure extensional regimes and recent geophysical data suggest the existence of a prolate ellipsoidal  
43 magma chamber below Stromboli. To constrain its volume we estimated the magma volumes  
44 associated with SO<sub>2</sub> degassing (during the 2007 major eruption) by applying a refined petrological  
45 model that allowed us to estimate the magma fluxes in the subvolcanic region (i.e., the magma flux  
46 entering the degassing zone). The long term trend of this magma flux follows an overall exponential  
47 decay, typical of pressurised magmatic systems, and indicated that magma rise was accompanied  
48 and followed by a slow decompression. This trend was shown to be consistent release of elastic  
49 strain accumulated either by pressurisation of the rocks surrounding the magma reservoir, or by  
50 pressurisation of the magma itself, or both. By analyzing the reservoir elastic response during  
51 magma decompression, we found that the current Stromboli magma chamber volume may be  
52 adequately constrained within 1-2 km<sup>3</sup>.

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## 58 **Introduction**

59

60 The role of volatiles is crucial in controlling petrogenetic and volcanic processes. Among these the  
61 most tangible are, for example, partial melting in the source region of primitive magmas, and the  
62 dynamics of fragmentation processes in magma chambers and volcanic conduits. Gaseous phases  
63 modulate magma transfer toward the surface and are involved in active degassing when magma  
64 batches are momentarily stored in the subvolcanic region before reaching the surface. Recent  
65 experimental progresses in igneous petrology and physical volcanology gave us the opportunity to  
66 refine solubility models to define degassing trajectories, and to better understand vesiculation  
67 processes. However, open conduit volcanoes are natural laboratories to investigate how volatiles  
68 migrate and concentrate under dynamic conditions. Fluid phases are involved in pressurising  
69 reservoirs, rule magma rise and wall-rock fracturing during dike injection that may lead to effusive-  
70 explosive eruptions at active volcanoes. A unique case to study these phenomena is Stromboli  
71 volcano located in the Tyrrhenian sea.

72 Stromboli is a composite stratovolcano with a persistent open-system activity used as a reference  
73 case by volcanologists to classify minor to intermediate volcanic eruptions (e.g. Newhall and Self,  
74 1982). The role of fluids in modulating strombolian activity is fundamental since it is involved in  
75 the so called “gas puffing” when magma pulses reach the open vents. During these phases, the  
76 sudden outburst of gaseous phases induces minor explosions with the projection of scoria bombs,  
77 lapilli and ash that characterise the typically mild strombolian activity. In addition, more violent  
78 events, also known as “major explosions”, are triggered by portions of undegassed magma that  
79 reaches the upper part of the conduit and explosively releases the exsolving gases. However, more  
80 energetic paroxysmal explosions are strongly linked to fragmentation processes induced by a  
81 “nearly instantaneous” ascent of a fluid-saturated *slug* that migrates through the conduit and  
82 explosively expands in its upper part.

83 Stromboli rejuvenated the scientific interest with the onset of major eruptive cycle on December 28,  
84 2002: lava effusion from summit vents was followed, on December 30, by a composite slump down  
85 the northern part of the cone, generating a tsunami that damaged part of the village and propagated  
86 to the north-eastern coast of Sicily. A multidisciplinary effort was coordinated by Italian Civil  
87 Defence authorities to mitigate volcanic risk and the potential effects of tsunami waves in the west-  
88 central Mediterranean region. This approach gave the scientific community the opportunity to  
89 carefully monitor the last effusive cycle (from February 27 to April 2, 2007) that culminated in the  
90 paroxysmal explosion of March 15, 2007.

91 In this review paper we integrate geophysical and petrological data to refine current models on the  
92 inner plumbing system of Stromboli volcano in the attempt to better decode the role of volatiles  
93 during magma storage and ascent. We will focus our analyses on available experimental petrologic  
94 data, melt inclusions and minerochemical data. We thermodynamically calculated mineral-melt-  
95 fluid equilibria on selected reactions to refine thermobarometry and magma compressibility during  
96 decompression. This will allow us, by means of geophysical models retrieved by analysing SO<sub>2</sub>  
97 emissions, to constrain magma chamber volumes for better interpreting the eruption dynamics.

98

99 **Fig. 1**

100

101 **Geological Setting**

102 Stromboli is the north-eastern island of the Aeolian Arc and has been built on a rather thin  
103 continental crust of the Calabrian arc (Fig. 1). This crustal unit extends westward beneath Lipari  
104 and Vulcano and then vanishes below Marsili submarine volcano. Fragments of this crust (quartzite,  
105 tonalite and diorite) were found as xenoliths in the older lavas of Stromboli (cf., Laiolo & Cigolini  
106 2006; and references therein). The volcano is located onto the Stromboli-Panarea alignment: a NE-  
107 SW fault subsidiary of the Tindari-Letojanni fault that propagates NNW-SSE though Eastern Sicily  
108 below Etna volcano (Ventura *et al.*, 1999; De Astis *et al.*, 2003; Acocella *et al.*, 2009). The Aeolian

109 Islands were built within the last 1.3 my (Gillot & Keller 1993). Lavas and tephra are essentially  
110 subduction-related calcalkaline, HK-calcalkaline, shoshonitic and potassic rocks. Subduction of the  
111 Ionian plate beneath the Calabrian Arc and was accompanied by a regional uplift (0.5-0.7 my),  
112 associated with crustal extension. Uplift within the forearc region was related to the rebound of the  
113 upper plate (Calabrian Arc and part of the Ionian lithosphere; e.g., Gvirtzman & Nur 2001). Current  
114 subduction is responsible of regional seismicity and volcanism (Fig. 1). A recent systematic  
115 analysis of teleseismic data by Pontevivo & Panza (2006) support the idea that the Moho below  
116 Stromboli is shallow, and located at about 15-17 km depth. These authors identified a low velocity  
117 layer ( $v_s \sim 3.5 \text{ km} \cdot \text{sec}^{-1}$ ) at a depth of 15-32 km below the cone. However, the source region of  
118 Stromboli primitive melts is well deeper, since primitive magmas likely originate in the mantle  
119 wedge above the Benioff Zone (e.g. Peccerillo, 2001; Schiavi *et al.*, 2012). Earthquake's  
120 subduction-related hypocenters reach a depth of approximately 500 km and cluster at about 350-150  
121 km below Stromboli (Fig. 1, see the INGV data bank ISIDE: Italian Seismological Instrumental and  
122 Parametric Data Base at <http://iside.rm.ingv.it/iside/standard/result.jsp?rst=1&page=EVENTS>).  
123 Recently, D'Auria *et al.* (2006) have located, by means of broadband network operative at  
124 Stromboli, few seismic events at depths 4.5-6 km below the summit of the volcano. These  
125 hypocenters are positioned in the surroundings of the top of the magma reservoir.

126

## 127 **Stromboli volcano**

128 The cone of Stromboli is approximately 3 km high with the top reaching 924 m a.s.l. (above sea  
129 level). The typical Strombolian activity is characterised by the persistent moderate explosions with  
130 the ejection of scoriae, lapilli, ash and bombs from vents positioned at the summit of Sciara del  
131 Fuoco (a collapsed sector delimited by a horseshoe-shaped scarp opening north-westward, e.g.  
132 Tibaldi *et al.*, 2009). The “mild” and persistent strombolian activity may be interrupted lava  
133 effusions, major explosions and paroxysms (Barberi *et al.*, 1993; Bertagnini *et al.*, 2011). A detailed  
134 study on the thermal regimes occurring at Stromboli, correlated with changes in volcanic activity

for over a decade, has been reported by Coppola *et al.* (2012). The material erupted during major and paroxysmal explosions consists of high vesicular lapilli and bombs, that are formed by the mingling of dark-coloured and crystal-rich scoria with golden-coloured crystal-poor *pumice* within a subvolcanic chamber (e.g., Francalanci *et al.*, 1999; Métrich *et al.*, 2001; 2005). The pumice contains olivine crystals that bear melt inclusions, likely representing the primitive shoshonitic Stromboli basalts. The April 5, 2003 and March 15, 2007 paroxysmal explosions occurred during both the last two effusive cycles. These explosions have been interpreted as a result of decompression induced by rapid magma discharge that triggered the ascent of a gas-rich deep-seated magma, leading to fragmentation of the pumiceous foam within the open conduit (Ripepe *et al.*, 2005). According to Cigolini *et al.* (2008) this process follows the elastic rebound of the chamber walls that will squeeze up portions of the undegassed magma column (that, in turn, would instantaneously rise and cross-cut the depth where critical gas saturation occurs), leading to magma fragmentation with the ejection of the “primitive golden pumices”. The amount of lava discharged during both effusive cycles may be estimated to be around  $1\text{--}1.3 \times 10^7 \text{ m}^3$  (Calvari *et al.*, 2005) and  $0.7\text{--}1 \times 10^7 \text{ m}^3$  (Neri & Lanzafame, 2009; Marsella *et al.*, 2009; Calvari *et al.*, 2010) (Fig. 2). Flank slumping onto Sciara del Fuoco occurred during several recent lava effusions (namely in 1879, 1916, 1919, 1930, 1944 and 1954, according to Barberi *et al.*, 1993) and generated tsunamis waves in this sector of the Mediterranean.

153

## 154 **Fig. 2**

155

The cone of Stromboli is subdivided into five units (Hornig-Kjarsgaard *et al.* 1993; Keller *et al.*, 1993; Francalanci *et al.*, 1993) of calc-alkaline type and shoshonitic affinity: three of them refer to the older part of the volcano, known as *Paleostromboli* (110–35 ky). The *Scari* lavas and tephra ( $34 \pm 3$  ky in age) are basalts and shoshonite that overlay the *Paleostromboli*. The *Vancori* lavas



(shoshonitic basalts and trachytes, with ages 26 to 13 ky) lay on top of the above sequence. The *Neostromboli* unit (13-6 ka) is in the northern sector of the island (Fig. 1b) and consists of shoshonitic and potassic lavas. Recent *San Bartolo lavas*, erupted during roman times (Arrighi *et al.*, 2004; Speranza *et al.*, 2008), are high-K calc-alkaline basalts with abundant mafic and ultramafic mantle xenoliths, ascribed to the roots-zone of Stromboli plumbing system (Laiolo & Cigolini 2006). Current lavas and tephra are high-K calc-alkaline basalts and shoshonites (cf., Francalanci *et al.*, 2004; Landi *et al.*, 2009; Pompilio *et al.*, 2012).

Structurally, the architecture of the island is the result of summit collapses of caldera type that took place from 100 to 13 ky BP (e.g., Tibaldi *et al.*, 2009). The last one led to the formation of Sciara del Fuoco (Fig. 1b). Recently, Marsella *et al.* (2012) reconstructed the morphological evolution of the Sciara del Fuoco since 1868, and correlated it with the on-going volcanic activity. Summit vents were persistently (and are currently) located along a N40°E normal fault. This trend is also consistent with the main direction of dyking (e.g., Tibaldi *et al.*, 2003; Ripepe *et al.*, 2005)

The structure of the subvolcanic plumbing system has been recently discussed. According to several authors (Métrich *et al.*, 2001, 2005; 2010), Bertagnini *et al.* (2003), Francalanci *et al.* (2004) the plumbing system is represented by a lower main chamber (that contains the primitive undegassed low porphyritic magma, LP magma, normally ejected as golden pumices) connected to an upper subsidiary reservoir where the more evolved magma is momentarily stored (degassed high porphyritic magma, HP magma, erupted as lavas and/or scorias during effusive and mild strombolian activity). According to Cigolini *et al.* (2008) the geometry of the plumbing system is strongly controlled by the local structural setting and stress distribution, and is simply represented by a prolate ellipsoid connected, in the subvolcanic region, with the active conduit. In this view the primitive LP magma is confined within its lower part, whereas the HP magma is located in its upper portion. However, geophysical data support the occurrence of an ellipsoidal structure below Stromboli volcano (Bonaccorso *et al.*, 2008).

186

## 187 **Analytical methods**

188 *Mineral phases* and *glasses* of samples collected at Stromboli during 2006 and 2007 were analyzed  
189 at the Dipartimento di Scienze della Terra of the University of Torino with a SEM Cambridge  
190 Instruments Stereoscan 360 equipped with an EDS Energy 200 and a Pentafet detector (Oxford  
191 Instruments). Operating conditions were 15 kV accelerating voltage, and a beam current of 2.68 nA.  
192 Quantitative data (with spot sizes of 2  $\mu\text{m}$  and 15  $\mu\text{m}$  in diameter, for minerals and glasses  
193 respectively) were obtained and processed using the Microanalysis Suite Issue 12, INCA (Suite  
194 version 4.01). Analyses were corrected for background, drift, mass absorption and secondary  
195 fluorescence by using natural and synthetic standards. The relative errors are within 2% for most of  
196 major elements and may go up to 10% for minor elements (with concentrations < 1.00 wt %).  
197 Bulk-rock compositions were acquired by inductively coupled plasma mass spectrometry (ICP-MS)  
198 analysis by ALS Chemex, Sevilla, Spain. Precision of analyzed major elements are better than  
199  $\pm 0.2\%$  for  $\text{SiO}_2$ , and  $\pm 0.1\%$  for the other oxides (with the exception of  $\text{MnO}$  and  $\text{P}_2\text{O}_5$  which show  
200 errors of ca.  $\pm 0.02\%$ ).

201

## 202 **Background Petrology**

203 We will briefly review the petrography of the lavas and the juvenile tephra erupted during 2002-  
204 2003 and 2007. The reader may refer to Landi *et al.* (2006; 2009), Cigolini *et al.* (2008), Métrich *et*  
205 *al.* (2001; 2005; 2010) and Bertagnini *et al.* (2003; 2008) for detailed descriptions.

206 *Lavas and scorias* erupted during the last two major eruptions (2002-2003 and 2007) are  
207 shoshonitic basalts with crystallinity up to 35-45% modal, consisting of diopsidic clinopyroxene  
208 (salite-augite), plagioclase and minor olivine on an interstitial glassy matrix (Fig. 3a; 3b).  
209 Groundmass microlites consist essentially of plagioclase and clinopyroxene. Scorias are slightly  
210 richer in  $\text{SiO}_2$  and alkali when compared to the pumices ejected during both paroxysmal explosions.  
211 *Basaltic pumices* have crystallinity <10% and consist of the above phases, with higher bulk  $\text{MgO}$

212 (Mg# 0.55-0.60) and CaO contents, whereas K<sub>2</sub>O is lower (2<wt %). Interstitial glass compositions  
213 are normally more evolved than those of the whole rock. *Melt inclusions* found within olivine  
214 crystals of the pumices are more primitive (with SiO<sub>2</sub> ranging between 45.6 and 49.4 wt % and Mg#  
215 ranging from 0.55 to 0.64). Some of these are richer in CaO and show lower alumina contents and  
216 are named *ultracalcic primitive melts*. These features have been ascribed by Pichavant *et al.* (2009)  
217 to crystal dissolution–reaction-mixing (DRM; cf., Danyushevsky *et al.*, 2004) of earlier fractionated  
218 clinopyroxene.

219

### 220 **Fig. 3**

221

222 Métrich *et al.* (2001; 2005; 2010) and Bertagnini *et al.* (2003; 2008), investigated in details  
223 primitive melt inclusions in olivine of the golden pumices. These contained 2-3.7 wt% H<sub>2</sub>O and  
224 700-1900 p.p.m. CO<sub>2</sub>. They suggest the primitive gas-rich shoshonitic, or high-K basalt, is residing  
225 within the middle-lower part of the reservoir and during its ascent and storage, in a subsidiary  
226 chamber, undergoes decompression-driven crystallisation leading to the formation of a crystal-  
227 richer magma batch. The ejected materials of this kind are lapilli and scoria bombs produced during  
228 the typically “mild and persistent” Strombolian activity. However, the undegassed primitive magma  
229 is stored at deeper levels and is solely erupted during paroxysmal explosions with the ejection of the  
230 “golden pumices”. According to the cited authors, mingling and interactions of dark-coloured and  
231 crystal-rich scoria with golden-coloured crystal-poor pumice normally occur during major violent  
232 explosions (e.g., Francalanci *et al.* 1999; Métrich *et al.* 2001; 2005; Landi *et al.* 2004).

233

### 234 **Fig. 4 and Table 1**

235

236 In Fig. 4 we report a summary of petrochemical data onto the  $K_2O$  vs.  $SiO_2$  diagram of Peccerillo &  
 237 Taylor (1976) that shows the variability of the erupted materials. The selected analyses of the  
 238 erupted products and melt inclusions are shown in Table 1.

239 Recently, Di Carlo *et al.* (2006) investigated phase relationships in melts of Stromboli golden  
 240 pumices by selecting a sample named PST 9, that represents the less evolved pumice in the dataset.  
 241 Experimental runs were carried out at temperatures of 1175-1050°C and pressures comprised  
 242 between 400 to 50 MPa for variable  $H_2O$  contents (5.5-1.2 wt %) and oxygen fugacities (within 2  
 243 log units above the Ni-NiO buffer). According to their results a diopsidic pyroxene is a liquidus  
 244 phase from 400 MPa to 150 MPa within the temperature range of 1175-1150°C. This phase is  
 245 followed by olivine eventually crystallising with pyroxene and plagioclase,  $\pm$  Fe-Ti oxide at lower  
 246 P-T regimes. This is basically consistent with the cooling experiments of Conte *et al.* (2006)  
 247 performed at 1 atm on somehow similar bulk compositions.

248 Di Carlo *et al.* (2006) experimentally confirmed that the magma type typical of the “golden  
 249 pumices” is an undegassed melt that, undergoing decompression, did not extensively crystallise  
 250 prior its ejection. Conversely, lava and scorias erupted during mild strombolian activity represent a  
 251 degassed magma that underwent a larger degree of crystallisation at lower pressure regimes.

252 More recently, Pichavant *et al.* (2009) performed additional experiments on PST9 from Stromboli  
 253 volcano under variable  $H_2O$  and  $CO_2$  concentrations. Experimental fluids coexisting with primitive  
 254 melts range from  $CO_2$  rich ( $X_{H_2O} \sim 0.2$ ) at 400 MPa to  $CO_2$  poor ( $X_{H_2O} \sim 0.8$ ) at 100 MPa. It was  
 255 found that clinopyroxene is the liquidus phase in all high pressure experiments, either under  
 256 hydrous or anhydrous conditions. In addition, at 0.1 MPa at temperatures of 1190 °C clinopyroxene  
 257 and Mg-olivine appear together on the liquidus, being indicative of the onset of a cotectic  
 258 relationship during decompression. The occurrence of this relationship was already established, for  
 259 the assemblages of the golden pumices, by Cigolini *et al.* (2008) who found nearly constant olivine-

260 clinopyroxene  $K_D^{ol-cpx}$  values (based on the  $Fe^{2+}$ - $Mg^{2+}$  exchange) in the natural assemblages  
261 (ranging  $0.5 \pm 0.06$ ).

262

### 263 *Stromboli fluids*

264 It is generally accepted that melting processes at subduction zones are generally controlled by water  
265 and carbon dioxide concentrations (e.g. Wyllie, 1979; Yoder, 1969; Mysen & Boettcher, 1975a;  
266 1975b). Indeed, these gases play a crucial role in most magmatic processes and their abundance has  
267 a direct influence on phase stabilities and crystal-melt-gas equilibria. In addition, gas exsolution  
268 during decompression substantially controls magma ascent and fragmentation processes. However,  
269 beside these, some other minor fluid phases play a considerable role as well. Among these, sulphur  
270 degassing is a key factor in decoding volcanic activity and magma dynamics in volcanic reservoirs.  
271 A cross check between the S related petrologic method and plume  $SO_2$  emissions is crucial for  
272 detecting magma recharge within the upper part of reservoirs, and to analyse eruption processes at  
273 active volcanoes.

274 Métrich *et al.* (2001; 2005; 2010) and Bertagnini *et al.* (2003) analysed sulphur, chlorine and  
275 fluorine within Stromboli glasses, including primitive melt inclusions. Sulphur concentrations  
276 found within the glasses of the “degassed” Stromboli scorias range from 170 to 250 p.p.m., whereas  
277 in primitive melt inclusions is comprised between 1200 and 2500 p.p.m.. Within the matrix glasses  
278 of porphyritic scorias, chlorine ranges from 1150 to 1270 p.p.m., while in the olivine-melt-  
279 inclusions is slightly higher, being comprised between 1500 and 2200 p.p.m.. Fluorine is rather  
280 variable: within scorias and pumices ranges from 780 to about 1050 p.p.m., whereas in primitive  
281 melt inclusions is somehow higher (960-1360 p.p.m.). Métrich *et al.* (2010) found that S is  
282 positively correlated with pressure, whereas Cl and F contents do not show appreciable variations  
283 with pressure changes. In particular, they suggested that  $SO_2$  is released at pressures of about 170  
284 MPa or lower. We will later discuss how sulphur degassing may give important information on  
285 magma volumes involved in recharge processes and magma chamber dynamics.

286 Stromboli fluids are also carefully monitored in volcano surveillance. Radon emissions (e.g.,  
 287 Cigolini *et al.*, 2009; 2013; Laiolo *et al.*, 2012) and CO<sub>2</sub> fluxes are automatically measured (Rizzo  
 288 *et al.*, 2009; Carapezza *et al.*, 2009). Carbon dioxide SO<sub>2</sub> and H<sub>2</sub>S are also continuously detected in  
 289 the volcanic plume by means of a multigas analyzer (Aiuppa *et al.*, 2005; 2009) and FTIR (La  
 290 Spina *et al.*, 2013). In addition, SO<sub>2</sub> is also carefully monitored by systematic COSPEC  
 291 measurements (Burton *et al.*, 2009). We will later consider some key issues that arise from the  
 292 analyses of the collected SO<sub>2</sub> timeseries when compared to eruption discharge rates.  
 293 In this section, we prefer to briefly review gas compositions sampled at the summit of Stromboli.  
 294 The data are few but quite significant. Following the early measurements of Tazieff and Tonani  
 295 (1963), gases released at Stromboli summit fumaroles were analysed by Carapezza & Federico  
 296 (2000) and by Finizola *et al.* (2003). Geothermometric estimates on the gases collected by Finizola  
 297 *et al.* (2003), obtained by means of the CO<sub>2</sub>-CH<sub>4</sub>-CO geothermometer (Chiodini & Cioni 1989),  
 298 give equilibration temperatures of 265-275°C. These temperatures are in good agreement with those  
 299 obtained graphically on the diagram  $\log(\text{H}_2\text{O}/\text{H}_2) + \log(\text{CO}/\text{CO}_2)$  vs.  $3\log(\text{CO}/\text{CO}_2) + \log(\text{CO}/\text{CH}_4)$   
 300 constructed by Chiodini & Marini (1998) that classify these fluids as superheated vapors. Thus,  
 301 these gases have essentially an hydrothermal origin (with  $X_{\text{H}_2\text{O}}$  approaching 0.90).  
 302 However, Martini *et al.* (1996) reported two gas compositions obtained on samples collected by M.  
 303 Ripepe within a fracture located at the rim of the NE summit crater. These gases have definitely  
 304 higher  $X_{\text{CO}_2}$  concentrations, ranging from 0.33 up to 0.73, and their sampling temperatures were 410  
 305 and 190°C. Noticeably, FTIR data reported by Burton *et al.* (2007) found that during the typical  
 306 strombolian explosions  $X_{\text{CO}_2}$  fractions are normally 0.33, and  $X_{\text{H}_2\text{O}}$  may be as high as 0.64.  
 307 Conversely, during quiescent degassing (or gas puffing) the above reach 0.13 and 0.83, respectively  
 308 (the rest, up to 1, being essentially SO<sub>2</sub> and HCl in equal proportions).  
 309  
 310  
 311

## 312 **Equilibrium assemblages in primitive melts**

313 Natural assemblages are complex systems. However, we may use experimental data and computer  
314 modelling (e.g., MELTS) to better constrain P-T-X variability. Both these approaches are based on  
315 the fact that equilibrium conditions are reached, and rule the evolution of the system. Unfortunately,  
316 the effects of volatile concentrations on phase stabilities are still inadequately known (such as the  
317 stability field of plagioclase and clinopyroxene in function of changing hydrous conditions, cf.  
318 Yoder, 1969; Kushiro, 1979; Métrich *et al.*, 2001). In this section, we will analyse the available data  
319 on Stromboli to identify equilibrium assemblages by using standard consolidated methods,  
320 normally applied in igneous petrology.

321 We thus explored the equilibrium conditions of Stromboli primitive basalts by analyzing  
322 minerochemical data and melts compositions in terms of distribution coefficients.

323 Among these, we first consider the olivine-liquid distribution coefficient  
324 ( $K_D^{ol-liq} = X_{Fe}^{ol} X_{Mg}^{liq} / X_{Mg}^{ol} X_{Fe}^{liq}$ ). According to Roeder & Emslie (1970), equilibrium conditions are  
325 satisfied for a value of 0.33 in this parameter. However, Kamenetsky *et al.* (1998) has suggested  
326 that this parameter may range from 0.27 to 0.29 in alkaline mafic magmas. According to Putirka  
327 (2008), the appropriate  $K_D^{ol-liq}$  values for basaltic magmas is 0.299 ( $\pm 0.05$ ). For Stromboli basalts,  
328 Métrich & Clocchiatti (1996) and Di Carlo *et al.* (2006) constrain the olivine-melt Mg-Fe  
329 distribution coefficient at 0.29 for equilibrium conditions. Thus a melt with a Mg-number (Mg#) of  
330 0.58 (i.e., one of the more mafic within the melt inclusions) will be in equilibrium with an olivine  
331 Fo<sub>83</sub>. The latter authors suggest that the Mg-Fe partition coefficient, for clinopyroxene-liquid  
332 equilibrium  $K_D^{cpx-liq} = X_{Fe}^{cpx} X_{Mg}^{liq} / X_{Mg}^{cpx} X_{Fe}^{liq}$ , is constrained to about 0.31 ( $\pm 0.06$ ) according to their  
333 experimental data. However, Putirka (2008) emphasised that the best estimate for this parameter is  
334 0.28 ( $\pm 0.08$ ) over a wide range of magma compositions.

335 Concerning the plagioclase-melt equilibrium, we considered a Ca-Na exchange between the two  
336 phases,  $K_D^{pl-liq} = X_{Ab}^{pl} X_{AlO1.5}^{liq} X_{CaO}^{liq} / X_{An}^{pl} X_{SiO2}^{liq} X_{NaO0.5}^{liq}$  accepting a value of 0.27  $\pm 0.05$  (cf. Putirka,

2008). In Table 2 we report a selection of representative mineral phases of the erupted products that are compatible with their coexisting melts and satisfy the above partitions coefficients.

## Table 2

An additional issue is plagioclase stability. First, petrography shows that plagioclase (locally euhedral) is an “ubiquitous” phase in all the erupted pumices. Second, Di Carlo *et al.* (2006) report a high-pressure run (at 400 MPa, 1100°C and 2.3 H<sub>2</sub>O wt% ) of a pumiceous melt (PST9) where plagioclase crystallises. Five other experimental runs at the same temperature, lower pressure and lower H<sub>2</sub>O contents define the plagioclase stability field. In the experimental works of Pichavant *et al.* (2009) plagioclase does not crystallise since they were performed at higher temperature (T>1150°C). However, starting from the same melt composition, Conte *et al.* (2006) found that plagioclase is a crystallising phase in their experiments at atmospheric pressure and temperature below the 1150°C. By testing the crystallisation Stromboli primitive basalts (including sample PST9) with the computer code MELTS (Ghiorso & Sack 1995), we found that clinopyroxene is liquidus phase above 250 MPa and 1175°C, whereas olivine becomes a liquidus phase together with clinopyroxene at lower pressure, and plagioclase would appear on the liquidus below 50 MPa for temperatures approaching 1100°C.

## Fig. 5

In order to assess the role of plagioclase during the crystallisation of Stromboli primitive basalts we analyzed the data by means of Pearce element ratios (Pearce & Stanley, 1991). These can be used to constrain the fractionating mineral phases within a given magma (cf. Russell & Nicholls, 1988). We thus plotted the data (ours plus those in literature) on the (2Ca+3Na)/K vs. Si/K diagram (Fig. 5a). A slope approaching to 1 indicates a pair removal. In our case the calculated slope is 0.90 (with a



correlation coefficient of 0.85), thus suggesting the fractionation of both phases (clinopyroxene and plagioclase). Fig. 5b with  $(2\text{Ca}+\text{Na})/\text{K}$  vs.  $\text{Al}/\text{K}$  indicates a higher proportion of clinopyroxene removal relative to plagioclase (0.60 to 0.40, respectively) (Fig. 5b). Finally, the  $2\text{Na}/\text{K}$  vs.  $\text{Al}/\text{K}$  diagram allow us to estimate that an average plagioclase of composition of about  $\text{An}_{82-76}$  was involved in the fractionation of Stromboli primitive basalts (Fig. 5c). Whereas a labradoritic plagioclase ( $\text{An}_{67-62}$ ) is involved in the crystallization of evolved melts. In both cases their values satisfy  $K_D^{pl-liq}$  constraints ( $0.23\pm0.01$ ) and are consistent with the petrography of lavas and scoriaceous materials.

## **Geothermometry and Geobarometry**

In this section we will first apply classical geothermometers and geobarometers to estimate the P-T regimes of Stromboli primitive melts. We will then refine our estimates by introducing a grid of selected reactions that will help us to further constrain equilibrium regimes for selected mineral-melt compositions. Finally, we synthesise our results and discuss them to better infer the extension of Stromboli magma reservoir.

First, the results obtained by systematically applying classical thermobarometers are reported in Table 3 (see the footnotes of this table relative uncertainties). Hydrous melt inclusions and bulk pumices with  $\text{H}_2\text{O}$  contents comprised between 2-3 wt %, show equilibration temperatures for their liquids in the range of 1100-1220°C. Lower values are obtained by using the algorithm of Sisson & Grove (1993), being constrained to 1100-1160°C. With the exception of the results obtained by applying the Putirka (2008) liquid thermometer (leading to temperatures of 1145-1185°C), other geothermometers substantially overlap, in the range of approximately 1160-1220°C.

Olivine-liquid geothermometers only partly overlap. Higher temperatures are obtained by using the one proposed by Beattie (1993) leading to temperatures of 1150-1220°C. The results obtained by calculating the Sisson & Grove (1993) and Putirka (2008) geothermometers are basically consistent

388 at lower temperatures ( $1100 \pm 30$  °C), whereas in the higher temperature ranges lead to about  
 389 1150°C and 1190°C, respectively (Table 3).

390

### 391 **Table 3**

392

393 The geothermometers that use clinopyroxene-liquid equilibria are essentially those proposed by  
 394 Putirka *et al.* (2003) and Putirka (2008; equations 32d, 33, 34), and practically overlap in the  
 395 temperature range of 1110-1220°C (Table 3). Finally, the clinopyroxene-liquid barometers  
 396 proposed by the latter author indentify rather variable equilibration pressures. Among these, the  
 397 most coherent with previous estimates (Métrich *et al.*, 2001; 2005; 2010; Bertagnini *et al.*, 2003 and  
 398 Cigolini *et al.*, 2008) are those obtained by applying his equation (30) of Putirka (2008), leading to  
 399 pressures of 170-360 MPa for the melt inclusions, and 210-290 MPa for the bulk pumices,  
 400 respectively. Unfortunately, Putirka's geobarometers show a good degree of uncertainty (see the  
 401 footnote of Table 3), so we had to find more reliable geobarometers.

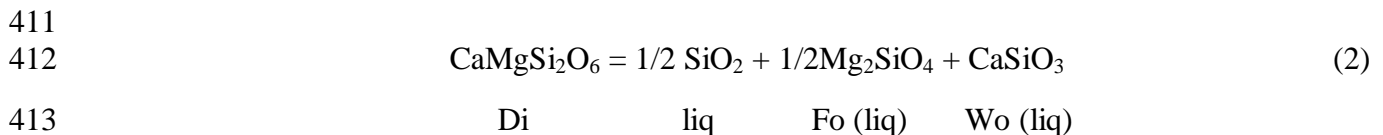
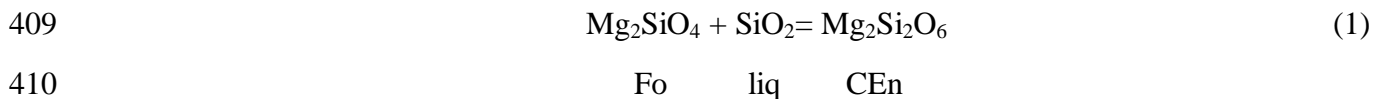
402

### 403 ***A grid of selected reactions***

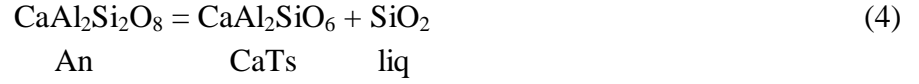
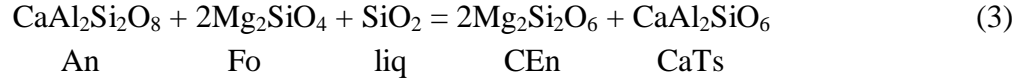
404 Thus, in order to minimize uncertainties related to the applications of the classical  
 405 thermobarometers treated above, we used a grid of selected reactions which involve solid-melt  
 406 equilibria to better constrain the P-T regimes that adequately describe our system.

407 These are the following:

408



414



Abbreviations below reaction components are: Fo – forsterite; CEn – clinoenstatite; Di – diopside; Wo – wollastonite; An – anorthite; CaTs – Ca-Tschermakite. Abbreviated mineral phases are solid, unless otherwise indicated (as liquid, liq)

Reactions (1) and (2) have been used to estimate P-T regimes for the coexistence of olivine and pyroxene, and to identify the upper stability field of diopsidic pyroxene in hydrous Stromboli basaltic melts. However, the use of melt components in thermodynamic calculations has the advantage of considering the water contents in the melt phase. In addition, reaction (3) is a rather efficient geobarometer (e.g., Laiolo and Cigolini 2006; Cigolini et al., 2008) in the stability field of bytownitic plagioclase and ferromagnesian phases consistent with the mineral assemblage found in Stromboli pumices. Finally, reaction (4) is typical of the low pressure assemblage where a labradoritic plagioclase is coexisting with augite and a differentiated groundmass melt under nearly anhydrous conditions (*ca.* 0.5 wt% H<sub>2</sub>O, cf. Stevenson & Blake 1998). Regarding this reaction, we will discuss the results of obtained in our previous study (Cigolini *et al.*, 2008). The degree of oxidation of the melt has been retrieved, following Kress & Carmichael (1991), from the Ni-NiO buffer of Hübner & Sato (1970), appropriate for Stromboli magma (cf., Laiolo & Cigolini 2006), taking into account the effect of pressure (in the light of the data of Robie *et al.*, 1979).

The Gibbs free energy for a generic *i* reaction is represented by

$$\Delta G_0^{P,T} = \Delta G_0^{1,T} + RT \ln K_i + \int_1^P \Delta V dP \quad (5)$$

444 where  $\Delta G_0^{1,T}$  is the Gibbs free energy for the reaction calculated at 1 bar and at the temperature of  
 445 interest ( $T$ ) by using the data of Berman (1988; cf. his Table 2, 3, and 4 p. 458-463) together with  
 446 those of Richet *et al.* (1982) for amorphous silica, and those of Ghiorso & Sack (1995) for other  
 447 melt components (Table A1, p. 209 of their work). A synopsis of used thermodynamic data is  
 448 reported in Table A2 of Cigolini *et al.* (2008).  $R$  is the gas constant and  $K_i$  is the equilibrium  
 449 constant for the  $i$  reaction considered, which may be calculated, knowing the chemical compositions  
 450 of the analysed phases, from the appropriate solution models. The last term is the difference of the  
 451 molar volumes of the reactants minus the products of the reaction integrated within the given  
 452 pressure range. This can be solved numerically at given equilibrium conditions by using the cited  
 453 data of Berman (1988), and those listed by Ghiorso & Sack (1995) for the melt components, that  
 454 include thermal expansion and compressibility.

455 In the studied reactions, activities for olivine, pyroxene and plagioclase were calculated according  
 456 to the solution models of Sack & Ghiorso (1989), Gasparik (1984; 1990) and Newton *et al.* (1980),  
 457 respectively. Activities of melt components were obtained according to the regular solution model  
 458 of Ghiorso & Sack (1995) and change with the water content in the melt phase. The solution of the  
 459 above equation has been obtained numerically, by minimizing the free energy  $\Delta G_0^{P,T}$  (that goes to  
 460 zero at equilibrium conditions) by fixing a given value of  $T$  and calculating  $P$  (or vice versa). The  
 461 same procedure has been applied to the set of the above reactions. A summary for the equilibrium  
 462 constants is given in Table 4. Melt compositions and mineral phases used in calculations are  
 463 summarised in Table 1 and Table 2, respectively.

464

#### 465 **Table 4**

466 The reference melt for reactions (1) and (2) are the selected melt inclusions found within the  
 467 olivines of ejected pumices (reported by Métrich *et al.*, 2001; 2005; 2010; and Bertagnini *et al.*,  
 468 2003) and identified as a primitive Stromboli basalts. Reaction (3) defines equilibrium conditions of

469 the golden pumices (bulk composition). Reference data are given in Table 1, whereas the selected  
470 equilibrium mineral phases are summarized in Table 2.

471 In the APPENDIX we report the thermodynamic solutions for the for melt inclusions (Tables A1  
472 and A2) and the bulk pumice (Table A3). The same data have been used to construct Figure 6.

473

## 474 **Fig. 6**

475

476 In particular, Stromboli primitive melts (Mg# 0.57 and CaO/Al<sub>2</sub>O<sub>3</sub> 0.73) are in equilibrium with a  
477 Fo-rich olivine (Fo<sub>84-80</sub>) and a diopsidic pyroxene (Wo<sub>44-48</sub>En<sub>43-48</sub>Fs<sub>6-9</sub>) at temperatures of 1140-  
478 1170 °C and pressures of 260 (±60) and 440 (±70) MPa for water concentrations ranging 2-4 wt%,  
479 respectively. More realistic best solutions are those for water being comprised between 2-3 wt%,  
480 giving temperatures of 1150-1160 °C and pressures of **280-340 MPa**. Similar reasoning for the Ca-  
481 rich melts (Mg# 0.63 and CaO/Al<sub>2</sub>O<sub>3</sub> 0.93) will give higher equilibration temperatures and lower  
482 pressures, with best solutions of 1160-1180 °C and pressures of 180 (±70) and 280 (±90) MPa with  
483 water contents of 2-3 wt%. The olivine in equilibrium with the ultracalcic melts are Fo<sub>88-90</sub>. Finally,  
484 equilibration P-T regimes for the golden pumices, given by the coexistence of the above mineral  
485 phases and a bytownitic plagioclase (An<sub>82-78</sub>) will constrain temperatures at 1120-1140 °C and  
486 pressures of 190 (±40) and 170 (±50) MPa for similar water contents (2-3 wt%).

487 Thermobarometric estimates for the upper part of the chamber were obtained by considering the  
488 equilibrium of phenocrystic rims with the coexisting groundmass melt (and the reader may refer to  
489 Cigolini *et al.*, 2008 for details). These data have been used, together with the above, to construct  
490 Fig. 7.

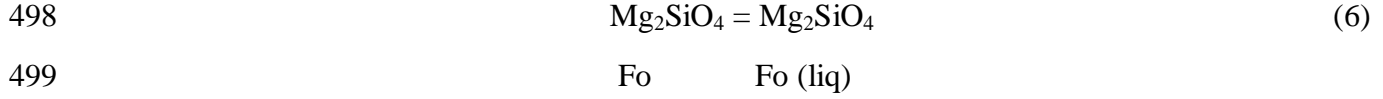
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## 492 ***Thermobarometry of Melt Inclusions***

493 Better constraining the P-T regimes of melts coexisting with volatiles is a key factor for decoding  
494 in-chamber magma dynamics (e.g., Witham, 2011a; Métrich *et al.*, 2011; Witham, 2011b).

495 In our earlier paper (Cigolini *et al.*, 2008) we calculated mineral-melt equilibria for the melt  
 496 inclusions reported by Métrich *et al.* (2001) according to the following reaction

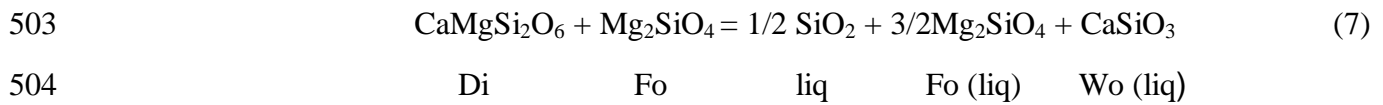
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500

501 By summing up the above with reaction (2), we obtain the following:

502



505

506 which is representative of the olivine-clinopyroxene cotectic relationship. According to Pichavant *et*  
 507 *al.* (2009) such a relationship is critical for assessing equilibrium conditions in Stromboli basalt.

508 We therefore extended our thermodynamic calculations by taking into account a more  
 509 comprehensive data set (including the data of Bertagnini *et al.*, 2003; Métrich *et al.*, 2001; 2005;  
 510 2010). In Table 5 we report the selected melts together with the appropriate clinopyroxene  
 511 compositions that satisfy the  $K_D^{Cpx-liq}$  requirements of Putirka (2008).

512

## 513 **Table 5 and Table 6**

514

515 In this section the notations  $X_{H_2O}^m$  and  $X_{CO_2}^m$  refer to the fractions of water and carbon dioxide in the  
 516 binary mixture dissolved in the melt under saturation conditions ( $X^m=X^V$ ): so that  $X_{H_2O}^m + X_{CO_2}^m = 1$ .

517 The above equilibrium can be solved for  $P$ ,  $T$ ,  $X_{H_2O}^m$  and  $X_{CO_2}^m$  for the given fluid concentrations (in  
 518 wt% as input data) by minimising the free energy of the above reaction and solving simultaneously  
 519 the solubility equations of Dixon & Stolper (1995) and Dixon (1997). The insertion of the molar

520 heat of solution for H<sub>2</sub>O in the melt (determined on albite melt by Silver & Stolper 1985) for  
 521 polythermal calculations has been shown to be negligible with respect to free energy minimization.  
 522 In other words, we are first solving thermodynamically the required thermodynamic conditions  
 523 along a Clausius-Clapeyron equation representative of minerals-melt equilibrium. The solution is  
 524 then found for the P-T conditions that satisfy the observed volatiles concentrations according to the  
 525 model of Dixon for alkaline basalts (Dixon, 1997). We trust that this approach is more reliable  
 526 because we are analyzing a three phase system (solids, melt and gas) that substantially improves the  
 527 commonly used two phase solubility models (e.g., Newman & Lowenstern 2002; Papale *et al.*,  
 528 2006).  
 529 In Table 6 we report the solutions for the above reaction for selected melts in equilibrium with the  
 530 chosen clinopyroxene compositions. These data indicate that primitive melt inclusions are in  
 531 equilibrium with ferromagnesian phases at pressures between 3.3 and 2 kbar and temperatures  
 532 ranging from about 1150 to 1200°C (with average values of about 1170°C, and only one melt  
 533 inclusion slightly above 1200°C). It is interesting to notice that calculated  $X_{CO_2}^m$  values range from  
 534 0.6 to 0.76. These estimates are in good agreement with the previous ones obtained by the models  
 535 of Newman & Lowenstern (2002) and Papale *et al.* (2006) models (e.g., Métrich *et al.*, 2005; 2010;  
 536 Bertagnini *et al.*, 2003). However, similar calculations performed by mean of reaction (6) for the  
 537 primitive melt inclusions lead to similar pressures but unrealistically higher temperatures (nearly  
 538 20-25°C higher, being incompatible with experimental data).  
 539 Notably, melt inclusions in scorias equilibrate with Fe-rich olivine (ca. Fo<sub>70</sub>) and an augitic  
 540 pyroxene at pressures of 60-100 MPa and temperatures ranging from 1120 to 1145°C (Table 7 and  
 541 Table 8). However, calculated  $X_{CO_2}^m$  are much higher (being comprised between 0.79 and 0.98)  
 542 since the water contents of these melt inclusions are very low (and substantially they are indicative  
 543 of a degassed magma). In this case also the thermodynamic solutions of reaction (6) give similar  
 544 pressure ranges as well, but considerable higher temperatures (up to 60°C) which are definitely

545 unrealistic. Therefore a cotectic relationship defined by reaction (7) is by far more appropriate for  
546 identifying solid-melt-fluid equilibration regimes in Stromboli basalts.

547

## 548 **Table 7 and Table 8**

549

### 550 *Summary of Thermobarometry*

551 Our best estimates suggest primitive hydrous melts preserved as melt inclusions in olivine (with  
552 2.3-3.8 wt% of H<sub>2</sub>O) are in equilibrium with forsteritic olivine and a diopsidic clinopyroxene at  
553 pressures of 200-340 MPa pressure and temperatures of 1150-1200°C. Average data of calculations  
554 on melt inclusions are 260 (±47) MPa and temperatures of 1170 (±17) °C. These regimes are  
555 representative of the base of the chamber, where the ferromagnesian phases may crystallise and  
556 cumulate (see Laiolo & Cigolini 2006; Cigolini et al., 2008). However, slightly more evolved melts,  
557 typical of the *golden pumices* (with 2-3 wt% H<sub>2</sub>O), are in equilibrium with Ca-pyroxene, forsteritic  
558 olivine and anorthitic plagioclase at 150–220 MPa and temperatures approaching 1120-1150°C.  
559 These materials likely represent the middle and main part of the chamber.

560 In turn, thermobarometric estimates for the upper part of the chamber were obtained by Cigolini *et*  
561 *al.* (2008) by considering the equilibrium of phenocrystic rims with coexisting groundmass melts  
562 (with Mg# of 0.20-0.25). The upper part of the chamber where the reservoir is connected with the  
563 conduit is confined at 140-120 MPa for temperatures approaching 1100°C (which is the measured  
564 effusion temperature for the lavas). Evolved melt inclusions in Fe-rich olivine (ca. Fo<sub>70</sub>) of the  
565 scorias show average equilibration pressures of 78 (±20) MPa, and temperatures 1138 (±14)°C,  
566 being indicative of crystallisation within the conduit or its root-zone that reaches the upper part of  
567 the chamber.

568 However, Stromboli magmas originate above the slab at an approximate depth of about 100 km  
569 (Schiavi *et al.*, 2012). From this depth melts will progressively migrate to upper levels and interact



570 with the ultramafic materials at depths of 34-24 km were temperatures reach 1200-1250°C (Cigolini  
571 *et al.*, 2008). In this region, the magma is percolating through the wallrocks of the LVZ and may  
572 converge into a feeder dyke that crosscuts the Moho. On their way to the surface, basaltic magmas  
573 may decompress along a steep adiabatic and/or isothermal curve crosscutting the stability field of  
574 diopside thus entering the liquidus field and generating ultracalcic melt inclusions. To better  
575 visualize this process we compiled summary of the mineral stabilities in Stromboli basalts (Fig. 7)  
576 in the light of the previously cited experimental data and the above equilibria, together with those  
577 reported by Cigolini *et al.* (2008) for the low pressure assemblage. The possible decompressional *P*-  
578 *T* path (at higher pressures) for explaining the origin of the ultracalcic melts is also plotted (upper  
579 path in Fig. 7). During decompression the magma path will cross the clinopyroxene stability field  
580 and enter the liquidus field. The earlier crystallized diopside is destabilized and will react with the  
581 melt leading to the formation of ultracalcic melts. Subsequent cooling associated with moderate  
582 decompression, and/or at nearly isobaric conditions, will lead to crystallization of the  
583 ferromagnesian phases again. This scenario is basically supporting the hypothesis of Pichavant *et*  
584 *al.* (2009) that suggest that Ca-rich melts are related to crystal dissolution–reaction-mixing (DRM;  
585 *cf.*, Danyushevsky *et al.*, 2004) of earlier fractionated clinopyroxene.

## 586 **Fig. 7**

587 Once the magma has entered the chamber, it will be stored undergoing fractionation and degassing.  
588 Further decompression and cooling, accompanied by crystallisation of clinopyroxene followed by  
589 Fe-richer olivine and plagioclase, will lead the basaltic magmas typical of scorias and lavas (Fig. 7).  
590 The most active part of the chamber is its axial sector, where the magma column, consisting of fresh  
591 magma of lower density and viscosity, will progressively migrate upward and degas before being  
592 erupted (*cf.*, Cigolini *et al.*, 2008).

## 593 **Geophysical constraints on magma chamber volumes: inference from SO<sub>2</sub> degassing**

### 594 *SO<sub>2</sub> flux and magma degassing rate*

595 Stromboli volcano is known for its remarkably steady state magma supply rates, as well as for the  
 596 continuous degassing rates associated with magma rise within the shallow magmatic system (Allard  
 597 et al., 1994). Recent SO<sub>2</sub> fluxes, representative of time-averaged sulphur emissions during  
 598 strombolian activity, were estimated to be approximately 150-200 t day<sup>-1</sup> (Burton *et al.*, 2007;  
 599 2009). However, the February–April 2007 eruption was accompanied by an “exceptional” (above  
 600 normal) degassing rate (Fig. 8) with mean SO<sub>2</sub> emissions of *ca.* 620 t day<sup>-1</sup>, i.e. 3-4 times higher  
 601 than those occurring during the typical strombolian activity (Burton *et al.* 2009). Although higher  
 602 than normal, the volume of magma degassed during the 35 days of lava effusion (February-April  
 603 2007), estimated from the SO<sub>2</sub> flux ( $\sim 2 \times 10^6$  m<sup>3</sup> dense rock equivalent, d.r.e., Burton *et al.*, 2009)  
 604 was drastically lower than the volume of erupted lava ( $\sim 8 \times 10^6$  m<sup>3</sup> d.r.e.; Calvari *et al.*, 2010,  
 605 Marsella *et al.*, 2009). Burton *et al.* (2009) suggested that such an unbalance could be reconciled by  
 606 considering an unrealistically high vesicularity of the 2007 lavas (>60%) or, better, by the assuming  
 607 that the most of the erupted magma volume (nearly  $6 \times 10^6$  m<sup>3</sup>) was already degassed when it was  
 608 erupted. Since, lava vesicularity at Stromboli is typically comprised between 16 and 32% (Fornaciai  
 609 *et al.*, 2009), the effusion of a large volume of degassed magma seems to be the case (due to the  
 610 drainage of a shallow plumbing system where the magma undergoes degassing). This may explain  
 611 the deficit in sulphur degassing observed during the February 2007 effusive eruption.  
 612 However, the long term record of sulphur output (Burton *et al.*, 2009) reveals that high values of  
 613 SO<sub>2</sub> flux persisted for several months after the ceasing of the effusive phase (see Fig. 8). In  
 614 particular, the trend shown in Fig. 8 suggests that the period of anomalous degassing (February to  
 615 October 2007) was characterised by an gradual decline of the SO<sub>2</sub> flux, which decreased to pre-  
 616 eruptive levels (i.e 150 t day<sup>-1</sup>; Burton *et al.*, 2009) only in October 2007, about 200 days after the  
 617 end of the lava effusion.  
 618 The sulphur output data shown in Fig. 8 allow us apply the petrologic method (cf., Palma *et al.*,  
 619 2011) in order to estimate the magma degassing rate ( $Q_{degas}$ ) of Stromboli during the period of  
 620 anomalous degassing (i.e. February to October 2007). In fact, the magma degassing rates (in terms

621 of the volumetric flux of magma undergoing sulphur degassing;  $\text{m}^3 \text{s}^{-1}$ ) can be derived by the  
 622 estimates of  $\text{SO}_2$  flux ( $\phi_{\text{SO}_2}$ ) and by the measurements of sulphur concentration in melt inclusions  
 623 (found within olivine phenocrysts) and in the interstitial glasses of the erupted materials (cf.  
 624 Kazahaya et al., 1994; Gerlach et al., 1996). To retrieve the magma degassing rates ( $Q_{\text{degas}}$ ), we may  
 625 write this relationship as:

$$626 \quad Q_{\text{degas}} = 10^2 \frac{M_s}{M_{\text{SO}_2}} \frac{\phi_{\text{SO}_2}}{\Delta S} \frac{1}{\rho_M} \quad (8)$$

627 where  $M_s$  and  $M_{\text{SO}_2}$  (both in g/mol) are the molecular mass of sulphur and sulphur dioxide,  
 628 respectively. The  $\phi_{\text{SO}_2}$  (kg/s) is the sulphur degassing rate and  $\Delta S$  (wt%) represents the total  
 629 outgassed sulphur, calculated as  $\Delta S = S_{mi}(1 - x_{mi}) - S_{ig}(1 - x_{ig})$ , where  $S_{mi}$  (wt%) and  $S_{ig}$  (wt%) are  
 630 respectively the sulphur concentration in the melt inclusions and in the interstitial glass, and  $x_{mi}$  and  
 631  $x_{ig}$  are, respectively, the crystals during the trapping of the melt inclusions (assumed to be zero) and  
 632 the crystals content in the matrix, as fraction. Moreover,  $\rho_M$  represents the bulk density of the  
 633 magma entering in the degassing zone.  $Q_{\text{degas}}$  was then calculated by taking into account a S content  
 634 of 0.24 wt% and 0.04 wt% in the primitive magma and in the scoria interstitial glasses, respectively  
 635 (cf. Bertagnini et al., 2003) and considering  $x_{ig} = 0.3$  (being consistent with Stevenson & Blake,  
 636 1998).

637 It is generally accepted that at Stromboli volcano, the magma degassing rate ( $Q_{\text{degas}}$ ) provides  
 638 constraints for the magma supply rate ( $Q_{in}$ ) or rather for the rate at which volatile-rich aphyric  
 639 basalt (LP magma) enters in the shallow plumbing system where sulphur degassing takes place (Allard et  
 640 al., 1994, 2008). Hence, the trend drawn by  $Q_{\text{degas}}$  (Fig. 8) can be regarded as the flux of magma  
 641 rising from a deep portion of the plumbing system (hereby defined as  $R_{\text{deep}}$ ), where the Low  
 642 Porphyrific (LP) magma resides with little crystallisation and degassing, to a shallowest storage  
 643 zone (defined as  $R_{\text{shallow}}$ ) where gas-melt separation occurs, thus promoting the steady strombolian  
 644 activity (Allard, 2008).

Thus, the magma supply (degassing) rate shown in Fig. 8 is likely representative of the LP magma flux that entered the degassing cell of Stromboli during and after the 2007 effusive eruption. As shown in Fig. 8 this magma flux see the contribution of two combined processes: (i) the constant supply of LP magma typical of the steady state regime of Stromboli ( $Q_{in-steady}$ ) and (ii) the anomalous (in excess) supply of LP magma entered in the degassing zone during and after an effusive phase ( $Q_{in-ex}$ ). Notably, the whole magma supply rate affected by degassing  $Q_{degas}$  (being equal to  $Q_{in-steady} + Q_{in-ex}$ ) describes a clear exponential trend (Fig. 8) that can be described by:

$$Q_{degas} = Q_{in-ex}^0 \cdot \left[ \exp(-t/\tau) + 1 \right] Q_{in-steady} \quad (9)$$

where  $Q_{in-ex}^0$  is the initial excess of magma supply (i.e., related to the effusive phase) and  $\tau$  is the calculated decay constant.

The coefficients retrieved from the best-fit regression indicates an initial  $Q_{in-ex}^0$  of  $0.73 \text{ m}^3 \text{ s}^{-1}$ , a decay constant ( $\tau$ ) of 82.5 days and a steady magma supply ( $Q_{in-steady}$ ) equal to  $0.1 \text{ m}^3 \text{ sec}^{-1}$ , the latter being in good agreement with the typical magma degassing rate of Stromboli (Allard *et al.*, 2008; Tamburello *et al.*, 2012). It follows that the integration of the curve plotted in Fig. 8 provides the cumulative volume LP magma transferred from  $R_{deep}$  to  $R_{shallow}$  during the anomalous  $\text{SO}_2$  flux (approximately 230 days). This volume is thus obtained by summing the volume of magma supplied at constant rate ( $V_{in-steady} = Q_{in-steady} \times 230 \text{ days} = \sim 2 \times 10^6 \text{ m}^3$ ) and the volume of magma in excess, obtained by integrating  $Q_{in-ex}$  ( $V_{in-ex} = Q_{in-ex}^0 \tau = \sim 5.2 \times 10^6 \text{ m}^3$ ). As a whole these calculations brings the total volume of the LP magma that entered the shallow reservoir ( $V_{in} = V_{in-ex} + V_{in-steady}$ ) to  $\sim 7.2 \times 10^6 \text{ m}^3$ . This value is surprisingly similar to the volume erupted during the 35 days of effusion ( $V_{erupt} \sim 8 \times 10^6 \text{ m}^3$  d.r.e. between 27 February and 4 April 2007) and implies that  $R_{shallow}$  was almost replenished in about 230 days. In addition it is remarkable to note that these results are in good agreement with the size of the degassing cell of Stromboli inferred by Allard *et al.* (2008;  $12 (\pm 3) \times 10^6 \text{ m}^3$ ), as well as with the maximum residence time of magma in the

degassing cell (being around 215 days, as assessed from the ratios of short-lived radioactive daughters; Gauthier *et al.*, 2000).

### ***Constraints on deep reservoir capacity and volume***

The fact that the long term trend of  $Q_{in-ex}$  leads to an overall exponential decay is intriguing, since this relationship is typical of pressurised magmatic systems and generally characterise the magma discharge during some basaltic eruption (Wadge, 1981). In the case of Stromboli we explain this behaviour by suggesting that the LP magma, rising and entering within  $R_{shallow}$ , was accompanied by a slow decompression of the deeper portion of the plumbing system of Stromboli ( $R_{deep}$ ). This process was slower than the one associated to lava effusion (that, in turn, has been lasting for 32 days; Bonaccorso *et al.*, 2008), and is indicative of the fact that deeper sector of the plumbing system exhibited a slow response to the pressure drop associated with lava effusion.

The exponential decline of  $Q$  after a short waxing phase is generally ascribed to the release of elastic strain accumulated either by pressurisation of the rocks surrounding the magma reservoir, or by pressurisation of the magma, or both (Wadge, 1981, Stasiuk *et al.*, 1993; Harris *et al.*, 2000, Aki & Ferrazzini, 2001; Huppert & Woods, 2002). According to these models, if the elastic strain is the sole driving force of magma, at any time  $t$ , the volumetric flux  $Q(t)$  is described by:

$$Q(t) = Q_0 \cdot \exp(-t/\tau) \quad (10)$$

where  $Q_0$  is initial flux and  $\tau$  is the time constant of the exponential decay. By analogy with a resistor-capacitor circuit, Aki & Ferrazzini (2001) proposed that the time constant  $\tau$  can be simply expressed as:

$$\tau = R_e C \quad (11)$$

693 where  $C$  is the capacity of the reservoir undergoing decompression (in  $\text{m}^3 \text{Pa}^{-1}$ ) and  $R_e$ , is the  
694 resistance of the “channel” (dike or conduit) to the magmatic flux (in  $\text{Pa s m}^{-3}$ ). Therefore, high  
695 reservoir’s capacity and/or high channel’s resistance will produce longer time constant and slower  
696 waning phases.

697 By assuming that the elastic strain is accumulated either by pressurisation of both the magma and  
698 the rocks surrounding the reservoir, the capacity  $C$  can be obtained, considering wall-rock and  
699 magma compressibility (cf., Mastin *et al.*, 2008), as

$$700 \quad C = \frac{\rho_M}{\rho_e} (\beta_R + \beta_M) V_R \quad (12)$$

701 where  $\rho_M$ ,  $\rho_e$  are the densities of magma and erupted lava, respectively,  $\beta_M$  is the magma  
702 compressibility,  $\beta_R$  is the wall-rock compressibility and  $V_R$  is the reservoir volume undergoing  
703 strain release. On the other hand the initial effusion rate  $Q_0$  is related to the channel (conduit)  
704 resistance  $R_e$  through the following Aki & Ferrazzini (2001):

$$705 \quad Q_0 = \frac{P_0}{R_e} \quad (13)$$

706 where  $P_0$  is the initial overpressure (in Pa). By integrating equation (9) it follows that a generic  
707 volume ( $V_{out}$ ) transferred outside the chamber is given by

$$708 \quad V_{out} = Q_0 \tau, \quad (14)$$

710  
711 or alternatively by

$$712 \quad V_{out} = CP_0 \quad (15)$$

713  
714 In order to constrain the volume of the deep reservoir undergoing the strain realise, the anomalous  
715 (in excess) supply of LP magma entered in the degassing zone ( $Q_{in-ex} = Q_{in-ex}^0 \cdot \exp(-t/\tau)$ ) can be  
716 analysed in the light of the above model. In particular, by determining the initial overpressure ( $P_0$ ),

717 magma compressibilities ( $\beta_M$  and  $\beta_R$ ) and densities ( $\rho_M$  and  $\rho_e$ ), the capacity ( $C$ ) and volume ( $V_R$ ) of  
 718 the reservoir can be constrained throughout equations (10) to (15).

719 During the onset of 2007 eruption a near instantaneous pressure drop of about 4-6 MPa was  
 720 transferred to the upper part of the reservoir that instantaneously decompressed (Calvari *et al.*  
 721 2011). At the same time, a large supply of CO<sub>2</sub>-rich and SO<sub>2</sub>-rich gas bubbles from the deep low  
 722 porphyritic (LP) sector of the reservoir (Burton *et al.*, 2009; Aiuppa *et al.*, 2010a, b), has been  
 723 invoked, as a consequence of depressurisation associated with lava drainage since the onset of the  
 724 eruption (27 February 2007; Bonaccorso *et al.*, 2008). Therefore, it is not unreasonable to assume  
 725 that an initial overpressure  $\geq 5$  MPa affected the whole plumbing system of Stromboli during the  
 726 beginning of the eruption. By setting  $P_0 = 5 - 10$  MPa and by assuming that the magma volume  
 727 entering in the degassing zone, was substantially the one removed outside the deeper reservoir  
 728 ( $V_{in-ex} \sim V_{out}$ , recalling that is equal to  $5.2 \times 10^6 \text{ m}^3$ ), we estimated a capacity  $C$  roughly comprised  
 729 between  $0.52$  and  $1.05 \text{ m}^3 \text{ Pa}^{-1}$  (equation 15).

730 We then calculated the variation of compressibility of primitive melts by taking into account the  
 731 variations in fluid contents during step-by-step decompression associated with bubble growth. Melt  
 732 densities ( $\rho_l$ ) were calculated according to Lange & Carmichael (1990), whereas the gas-mixture  
 733 densities ( $\rho_g$ ) were obtained by applying the Modified Redlich-Kwong (MRK) equation of  
 734 Holloway (1977). The quantities of water and CO<sub>2</sub> (in wt%) released during decompression were  
 735 obtained by applying the computer code VOLATILECALC of Newman & Lowenstern (2002). Thus,  
 736 the magma bulk density ( $\rho_M$ ) and compressibility ( $\beta_M$ ) were calculated according to Huppert and  
 737 Woods (2002, equations 5 and 6 of their work) that take into account crystallinity and the  
 738 contribution of wall-rock compressibility ( $\beta_R = 3^{-11} \text{ Pa}^{-1}$ ). The latter parameter was retrieved  
 739 (assuming a Poisson ratio of 0.25) from the rigidity calculated by D'Auria *et al.* (2006) for  
 740 earthquakes located in the surrounding of the top of the magma reservoir (at 4.5-6 km depth). The  
 741 selected melt for calculating the appropriate magma compressibility ( $\beta_M$ ) is the average of the

analyses of melt inclusions reported in Table 5. Thus, given the magma pressure-dependent compressibility model shown in Fig. 9, we may consider that at 200 MPa (i.e., the main portion of the reservoir) the magma compressibility,  $\beta_M$  reaches  $4^{-10} \text{ Pa}^{-1}$ , while the wall-rock compressibility,  $\beta_R$ , is almost one order of magnitude less. In Table A5 we report the isothermal decompression path together with the results of our calculations. Similarly, we may assume that during its transfer from the deep to the upper part of the chamber, the magma changed its density from  $\rho_M = 2690 \text{ kg m}^{-3}$  (unvesiculated magma) to  $\rho_e = 2480 \text{ kg m}^{-3}$  (erupted vesiculated magma). By using equation (12), for the above calculated reservoir capacity ( $C = 0.52$  and  $1.05 \text{ m}^3 \text{ Pa}^{-1}$ ), we estimated the volume of the reservoir undergoing the elastic strain release ( $V_R$ ): being comprised between 1 and  $2 \text{ km}^3$ , in good agreement with the lower volume estimates of Cigolini *et al.* (2008) for the Stromboli magma chamber.

753

## 754 **Fig. 9**

755

## 756 **Discussion and conclusions**

In this contribution we have revisited the data on the last two major eruptions at Stromboli volcano and concentrated our analysis on the 2007 eruption. Our main goal was to offer a comprehensive view of the problems connected with the interpretation of the onset and evolution of these events and how their analysis may contribute to better understand the inner dynamics of this complex volcanic system. We therefore focussed our research on how volatiles contents affect solid-melt equilibria and how they behave during magma storage and decompression. These processes have a direct link on the type of volcanic activity that is taking place at Stromboli. However, our effort was to reconcile the large amount of data collected on this volcano with a plausible picture that explains how this volcano works. The first part of this review paper was concentrated in revisiting mineral phase stabilities in Stromboli basalt and how these, together with volatiles, may affect magma



compressibility during progressive decompression. In this perspective, one of the open issue we attempted to unravel is the effective shape and extension of Stromboli magma chamber. In general, the geometry and dynamics of magma chambers are still debated issues (e.g., Bonafede *et al.*, 1986; Dragoni & Magnanesi, 1989; Jellinek & De Paolo, 2003). Unfortunately, these authors essentially model magma chamber as spherical objects. In a recent contribution Zellmer & Annen (2008) discussed a full variety of plumbing systems, and attempted to correlate their geometry with magma transfer rates and magmatic differentiation processes. For instance, Stromboli is classified within the field of relatively small reservoirs found within extensional (or transtensional) settings, where the magma is stored for  $10^3$  years at the most. However, in a recent paper Mastin *et al.* (2008) suggested a possible shape for Mount Saint Helen magma chamber that may be regarded as a prolate ellipsoid. Moreover, Gudmundsson (2006) discussed the possible shapes of magma chambers in relation to the structural setting of the region and indicates a prolate ellipsoid as a recurrent shape. In our specific case we are dealing with a rather unique case where seismic data indicate that the major structure underneath Stromboli is essentially a normal fault (the Stromboli-Panarea alignment) with a minor right-lateral strike-slip component SW of Panarea (e.g., Caccamo *et al.*, 1996). According to Acocella *et al.* (2009) in the Stromboli area stress distribution approach pure extensional regimes, being consistent with the earlier works of Tibaldi (2001; 2004) and Tibaldi *et al.* (2003). Following Shaw (1980) and assuming that stress regimes are proportional to the geometry of the reservoir (whose vertical extension can inferred by fine thermobarometry), we may infer stress distribution and give more reliable estimates than those proposed earlier by Cigolini *et al.* (2008) for the Stromboli magma chamber volume. Another open issue is how the system pressurizes and what factors are involved in the development of this process. Allard *et al.* (2008) have shown that the eruption rate during mild and persistent Strombolian activity is at most  $0.02 \text{ m}^3/\text{sec}$ , whereas input flow rates at depth may be as high as  $0.3\text{-}0.6 \text{ m}^3/\text{sec}$ . This mechanism will progressively pressurize the “open” plumbing system until a critical threshold is reached and a major eruptive cycle begins. In this context strain rates at the chamber walls seems to be a critical

793 factor that need to be investigated in details and could be one of the goals for future research  
794 advancements.

795 In conclusion, current results can be summarised as follows:

796 i. The role of volatiles has been shown to be critical for identifying the equilibrium  
797 conditions of Stromboli magma during magma storage and decompression.

798 ii. Thermobarometric estimates indicate that Stromboli primitive hydrous basaltic melts  
799 (preserved as melt inclusions in olivine with 2-3.7 wt% H<sub>2</sub>O) are in equilibrium with  
800 forsteritic olivine and diopside at pressures of 200-340 MPa pressure and temperatures of  
801 1150-1200°C for  $X_{CO_2}^m$  comprised between 0.6 and 0.76 (which is quite similar to the  
802 magmatic gas composition sampled at the crater rim). The higher pressure regimes are  
803 representative of the base of the chamber, where the ferromagnesian phases may  
804 crystallise and cumulate. A particular case is given by Ca-rich or ultracalcic melts that are  
805 regarded as a result of decompression along a steep adiabatic and/or isothermal curve.  
806 During this process the magma will crosscut the stability field of diopside and enter the  
807 liquidus field. The earlier crystallized diopside is destabilized and will react with the melt  
808 leading to the formation of *ultracalcic melts*. Pichavant et al. (2009) ealier discussed this  
809 possibility and we hereby explain how this process is occurring and quantify the P-T  
810 regimes associated with this dynamics.

811 iii. Ejected golden pumices (with 2-3 wt% H<sub>2</sub>O), are in equilibrium with Ca-pyroxene,  
812 forsteritic olivine and anorthitic plagioclase at 150–220 MPa and temperatures  
813 approaching 1120-1150°C. These materials likely represent the middle and main part of  
814 the chamber.

815 iv. Evolved melt inclusions in Fe-richer olivine (ca. Fo<sub>70</sub>) of the scorias show average  
816 equilibration pressures of 78 (± 20) MPa, and temperatures 1138 (± 14)°C, being

indicative of crystallisation within the conduit or its root-zone that reaches the upper part of the chamber.

v. The lower part of the chamber is located at 11-13 km below the summit whereas the upper part of the chamber reaches 4.5-5 km below the summit itself;

vi. the magma chamber is substantially zoned and the undegassed magma is stored below the latter depth (and may be ejected as pumice during paroxysmal explosions and, more rarely, major explosions);

vii. during its storage, within the upper part of the chamber and within the dike feeding the summit craters, the magma undergoes progressive degassing, cooling and crystallisation. These processes are involved in the genesis of lavas and scorias (that represent the degassed magma reaching the surface).

viii. The Stromboli plumbing system consists of an ellipsoidal magma chamber of 1-2 km<sup>3</sup> in volume, likely a prolate ellipsoid whose upper part is connected with the feeder dike.

ix. during major eruptions associated with lava effusions the dike feeding the summit vents is substantially rejuvenated and the upper part of the chamber is progressively replenished by the undegassed magma. During the 2007 eruption, the original magmatic equilibrium was restored in about 230 days from the beginning of the effusive activity (as inferred by our analysis on SO<sub>2</sub>-derived magma fluxes).

## Acknowledgements

The research was funded by the University of Torino - Fondazione Compagnia di San Paolo. Additional funds were provided by MIUR and made available through the Cooperative Project among the Departments of Earth Sciences of the University of Torino and the University of Firenze (in the framework of the Devnet Project, funded by the Dipartimento della Protezione Civile, DPC). We are indebted to M. Ripepe and M. Pichavant for the stimulating discussions. We also thank Sébastien Jego and an anonymous reviewer for revising an earlier draft of the paper. Special

843 acknowledgements to INGV-Catania for making the SO<sub>2</sub> data available on the web. Additional  
844 funds for the improving our computing hardware were provided by Fondazione Cassa di Risparmio  
845 di Torino..

846

## 847 **APPENDIX**

848 We hereby report Table A1, A2 and A3 that summarizes the results of thermodynamic calculations  
849 regarding reactions (1) (2) and (3) that enabled us to construct Fig. 6.

850

### 851 **Table A1, Table A2, Table A3**

852

853 In Table A4 we summarize the list of symbols used in the equations discussed in the text.

854

### 855 **Table A4**

856

857 In Table A5 we report step-by-step calculations for the isothermal decompression of a primitive  
858 Stromboli basalt (cf., Fig. 9).

859

### 860 **Table A5**

861

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863

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1334 **Figure Captions**

1335

1336 **Fig. 1.** Sketch of the Aeolian Islands with the major tectonic units of the region (modified after De  
1337 Astis *et al.*, 2003; Ventura *et al.*, 2013). **(a)** Stromboli is located on the Stromboli-Panarea  
1338 alignment, a normal fault associated with pure extensional regimes (Acocella *et al.*, 2009). Insets  
1339 show the crust-mantle structure along a WNW-ESE cross section (cf. Pontevivo & Panza, 2006;  
1340 Corazzato *et al.*, 2008), and the summary of earthquake locations and magnitudes below the  
1341 Aeolian arc. The earthquakes reported are included within a sector with 37°-40° latitude, and 12°-  
1342 15° longitude, respectively (see text for details). LVZ: low velocity zone;  
1343 **b)** simplified geological map of Stromboli with major collapses and faults (modified after Hornig-  
1344 Kjarsgaard *et al.*, 1993; Tibaldi *et al.*, 2009).

1345

1346 **Fig. 2.** Selected images of Stromboli volcano during and following the 2007 major eruption; **(a)** the  
1347 2007 lava field emplaced onto earlier lavas and debris of Sciara del Fuoco. Picture taken on August  
1348 3, 2007 during a helicopter survey; **(b)** view of the summit crater area where the typical strombolian  
1349 activity was resumed on July 2, 2007. Summit vents are located on the N40°E graben-like fracture  
1350 zone (red dashed line). Picture taken on August 3, 2007 during a helicopter survey.

1351

1352 **Fig. 3.** Microphotographs of the petrographic and textural features of Stromboli lavas, scorias and  
1353 pumices erupted in 2002-2003; **a)** *scoria* erupted on February 16, 2007 with euhedral phenocrysts  
1354 of plagioclase, and clinopyroxene on a glassy matrix. Vesicles are also present (white). Scorias are

1355 interpreted as “conduit assemblages”. Plane polarized light; **b)** microphotograph of lava erupted in  
 1356 March, 2007 during the last effusive event. Lava is phenocrysts-rich (around 40%) mainly  
 1357 represented by clinopyroxene and plagioclase, with the typical oscillatory zoning. Vesiculation is  
 1358 relatively high. Plane polarized light; **c)** Photomicrograph of a quenched “*golden pumice*” erupted  
 1359 during the paroxysmal explosion of March 15, 2007. Vesicularity is very high (~70%) with bubbles  
 1360 of variable size and low degree of cristallinity (<10%) with microliths of plagioclase, clinopyroxene  
 1361 and olivine (not visible at this scale). Plane polarized light.

1362

1363 **Fig. 4.** Low silica-moderate K<sub>2</sub>O sector of the diagram of Peccerillo and Taylor (1976) with the  
 1364 plotting of the available data on the materials erupted at Stromboli; **a)** lavas and scorias, including  
 1365 matrix glasses. Grey fields refer to historical materials erupted from Stromboli volcano since 1906  
 1366 (these data were summarized by Francalanci *et al.*, 2004; Pompilio *et al.*, 2012). The dark grey field  
 1367 show the glass composition of the 2002-2003 and 2007 erupted materials (data reported by Landi *et*  
 1368 *al.*, 2006; 2009) ; **b)** pumices erupted during paroxysmal and major explosions; melt inclusions are  
 1369 those reported by Métrich *et al.* (2001; 2005; 2010) Bertagnini *et al.* (2003). The grey field shows  
 1370 the bulk composition of the hystorical pumices reported by Pompilio *et al.* (2012).

1371

1372 **Fig. 5.** Pearce element ratio plots for Stromboli materials (including melt inclusions in olivines of  
 1373 the pumices); **a)** 2Ca+3Na/K versus Si/K diagram indicates fractionation of clinopyroxene and  
 1374 plagioclase; **b)** the diagram 2Ca+3Na/K versus Al/K indicates a lower proportion of plagioclase  
 1375 removal relative to pyroxene (0.40 to 0.60, respectively); **c)** Pearce element 2Na/K versus Al/K  
 1376 showing that fractionation of a bytownitic plagioclase (An<sub>82-76</sub>) has been effective in primitive melts  
 1377 (the regression line is  $y=0.276x+0.5312$  with a correlation coefficient  $r=0.85$ ); in more evolved  
 1378 melts the fractionating plagioclase is a labradorite (An<sub>67-62</sub>) (in this case the regression line is  
 1379  $y=0.3576x+0.313$  with a correlation coefficient  $r=0.74$ ). Reference curves for fractionation of  
 1380 anorthite (An<sub>100</sub>) and albite (An<sub>0</sub>) are also reported for comparison.



1381

1382 **Fig. 6.** Thermobarometry of melt inclusions in olivine in primitive Stromboli basalts and pumices  
1383 erupted at Stromboli. **(a)** *P-T* estimates for the equilibrium crystallization of primitive Stromboli  
1384 basalts (preserved in melt inclusions). The graph summarizes the *P-T* grid obtained by solving  
1385 thermodynamically reactions (1) and (2) indicated by the large numbers in parentheses. Smaller  
1386 numbers are the water contents within the melt (wt% H<sub>2</sub>O). Best solutions are outlined by  
1387 rectangles (see text and Table A1). **(b)** *P-T* estimates for ultracalcic melt inclusions following the  
1388 same procedure indicated above. Note that these melts equilibrate at moderately higher  
1389 temperatures and slightly lower pressures than these reported above (Fig. 6b, Table A2).  
1390 **(c)** *P-T* estimates for the crystallisation of the mineral phases of the pumices. In this case best  
1391 estimates are obtained by solving thermodynamically reaction (2) and (3). See text and Table A3.

1392

1393 **Fig. 7.** Schematic sketch of phase stabilities for Stromboli primitive basalt and suggested *P-T* paths  
1394 during decompression. Phase stabilities are consistent with previously discussed equilibria and the  
1395 experimental data of Di Carlo *et al.* (2006), Pichavant *et al.*, 2009. The stability field of the low  
1396 pressure assemblage has been reconstructed following Cigolini *et al.* (2008) and the experimental  
1397 work of Conte *et al.* (2006). Average primitive melt inclusions and ultracalcic melts are represented  
1398 by white and grey squares, respectively (with their relative standard deviations, see Tables 5 and 6,  
1399 see text). The average of evolved melt inclusions found in Fo<sub>70</sub> of the scorias are represented by a  
1400 white circle (see Table 7 and 8). Line (a) represents the final decompression of the evolved  
1401 Stromboli basalt erupted as scoria or lava; line (b) is the possible final *P-T* path associated with the  
1402 ejection of pumices during paroxysmal explosions.

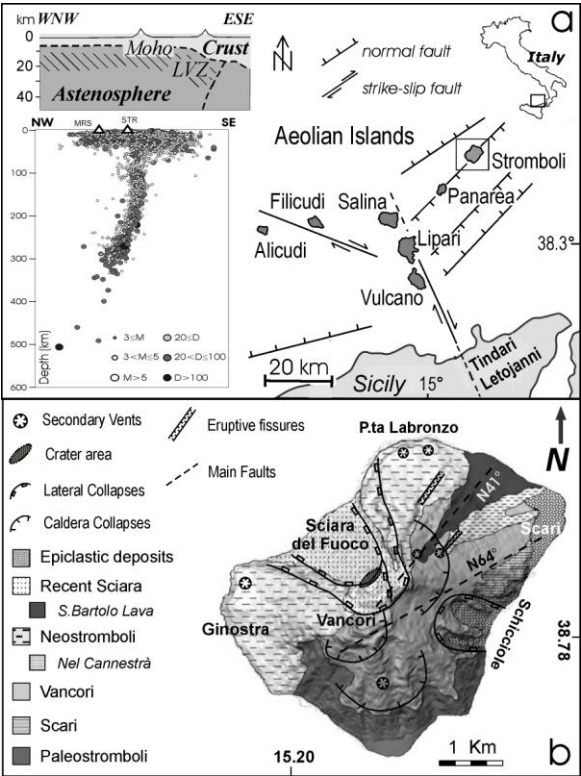
1403

1404 **Fig. 8.** Sulphur degassing rate (SO<sub>2</sub> flux - left axis) recorded between 27 February 2007 (the onset  
1405 of the effusive activity) and 15 October 2007. This period was characterized by anomalous  
1406 degassing rates, that exceeded average values recorded during the typical strombolian activity

(~100 ton/day). The SO<sub>2</sub> flux data are from INGV, available at [http://www.pa.ingv.it/comunicati/Stromboli/comunicati\\_stromboli.php](http://www.pa.ingv.it/comunicati/Stromboli/comunicati_stromboli.php). On the right axis, sulphur emissions were converted into volumetric flux of magma degassing ( $Q_{degas}$ ), by using the petrologic method (see text for details). The trend obtained was fitted by using an “exponential + offset” equation (dashed line), where the offset component represents to the magma supply rate during the typical strombolian activity ( $Q_{in,steady}$  – cf. bottom of the graph) and the exponential component represents to the “excess” of magma supply rate recorded during and after the 2007 effusive activity ( $Q_{in,ex}$  – stippled field). The exponential trend retrieved for  $Q_{in,ex}$  is ascribed to decompression from deep storage zone.

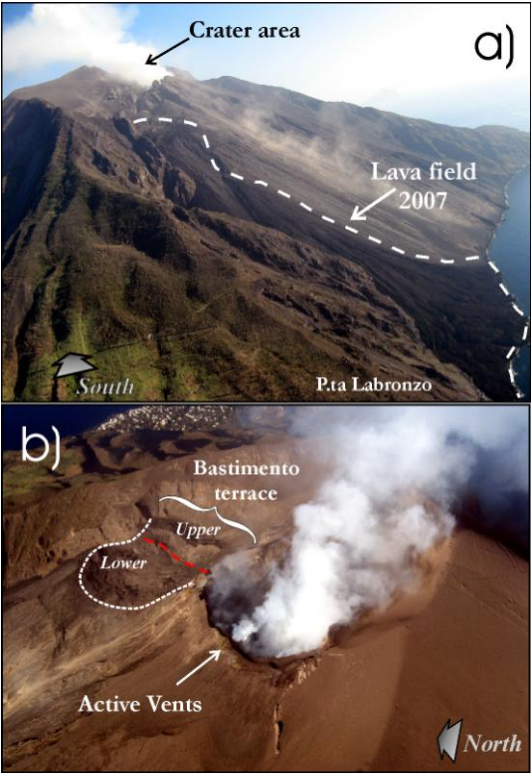
**Fig. 9.** Representative graphs that synthesize the isothermal decompression of a primitive Stromboli basalt (the selected melt is the average of melt inclusions reported in Table 5; see Appendix 5 for calculations). **(a)** Solubility of water and carbon dioxide as a function of pressure (for a temperature of 1170 °C) and **(b)** cumulative gas volume fraction during decompression calculated according to VOLATILECALC (Newman & Lowerstern 2002). **(c)** Variation of compressibility for the above primitive Stromboli basalt during decompression due to bubble growth and crystallization (see text for details).

1431 **Fig. 1**



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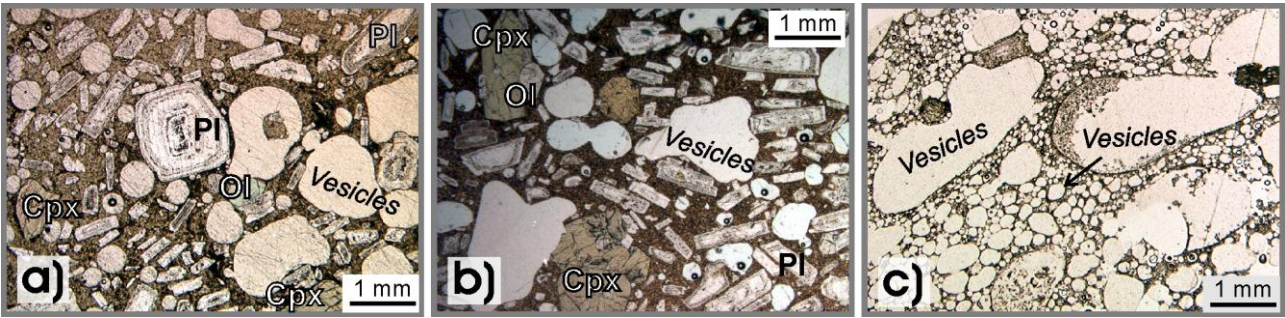
1433 **Fig. 2**



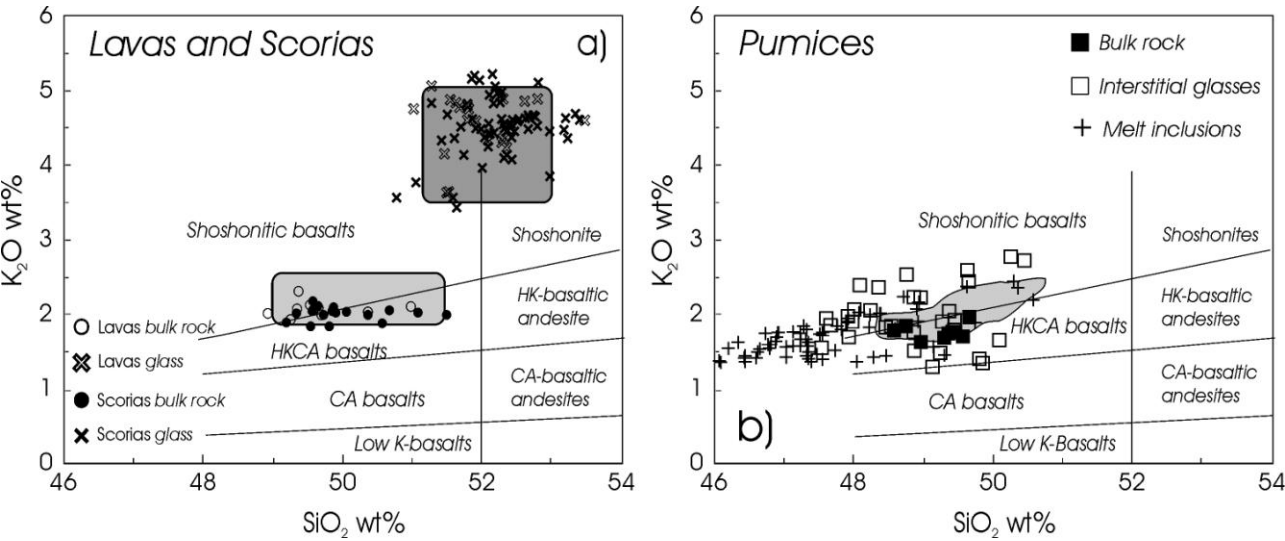
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1435 **Fig. 3**

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1437 **Fig. 4**



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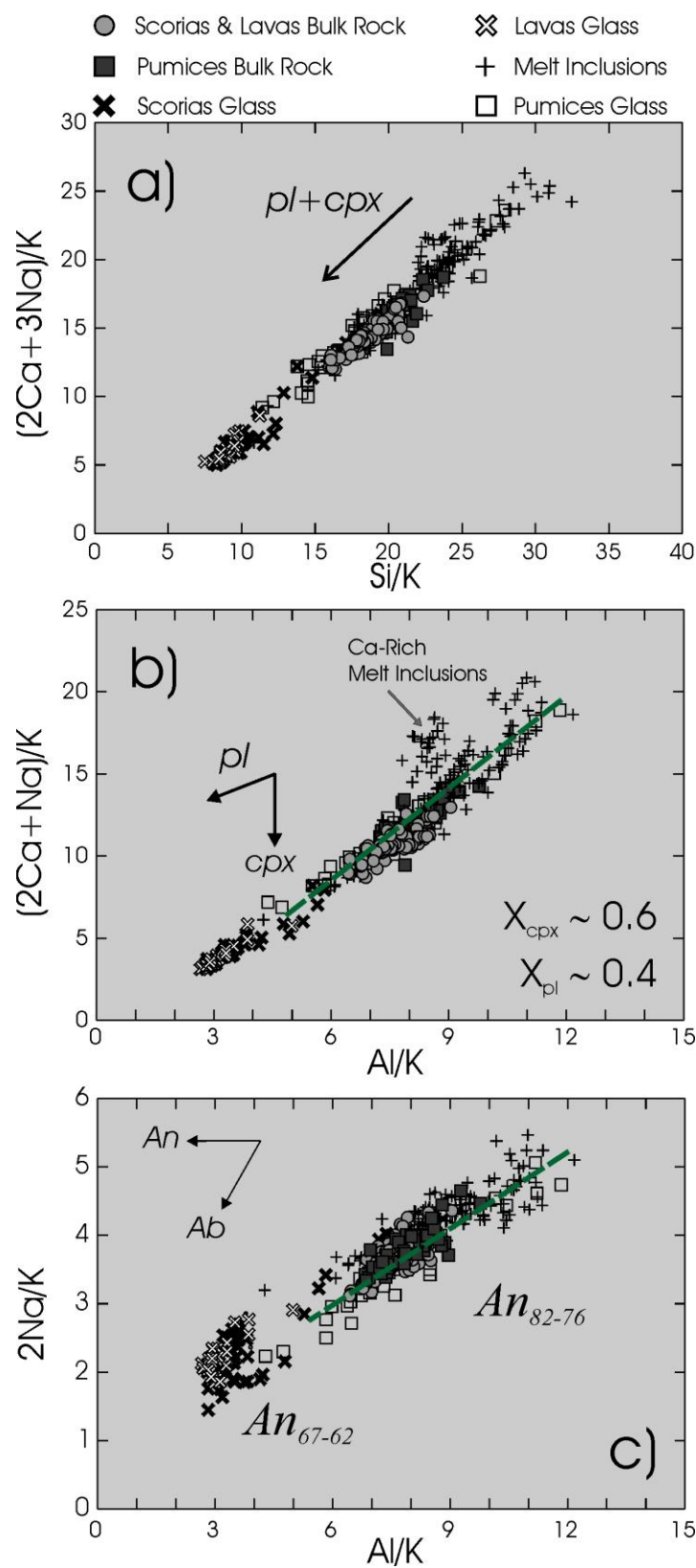
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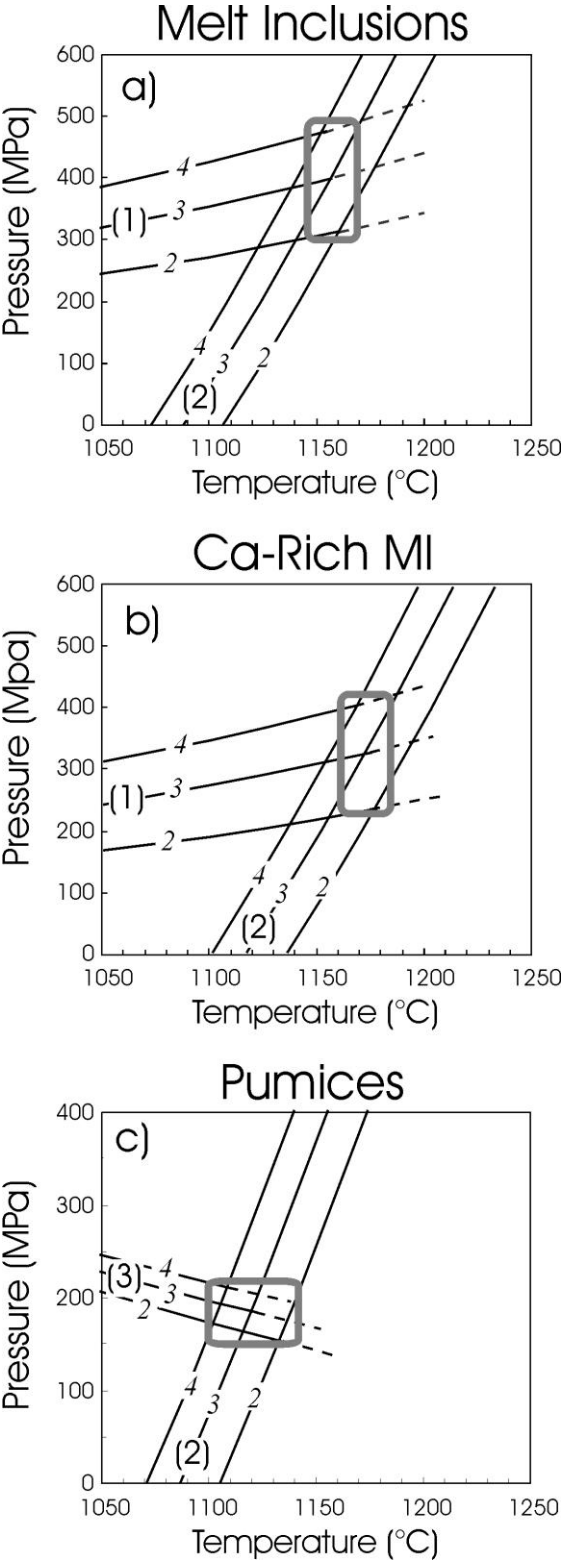
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1446 **Fig. 5**



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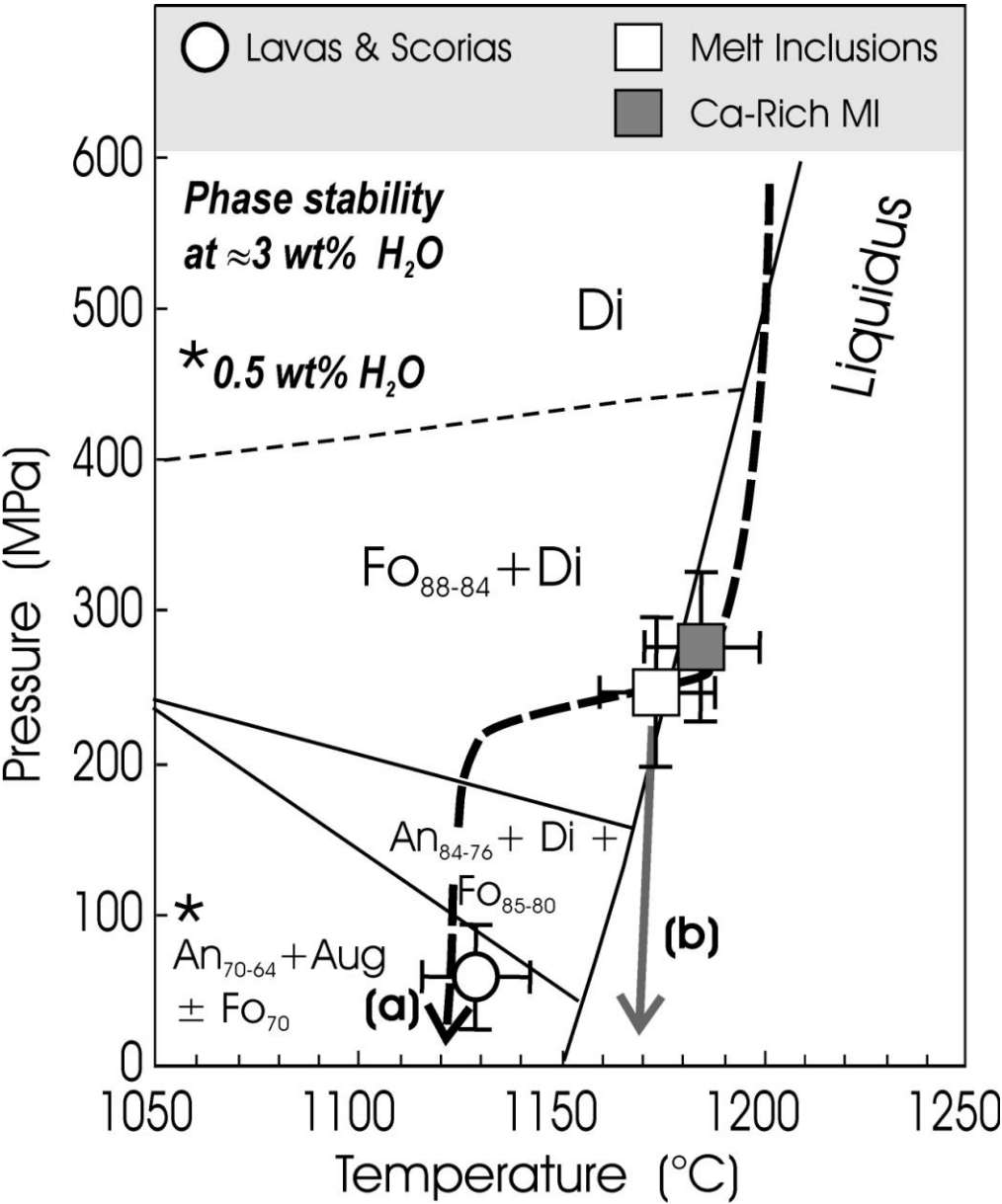


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1453 **Fig. 7**



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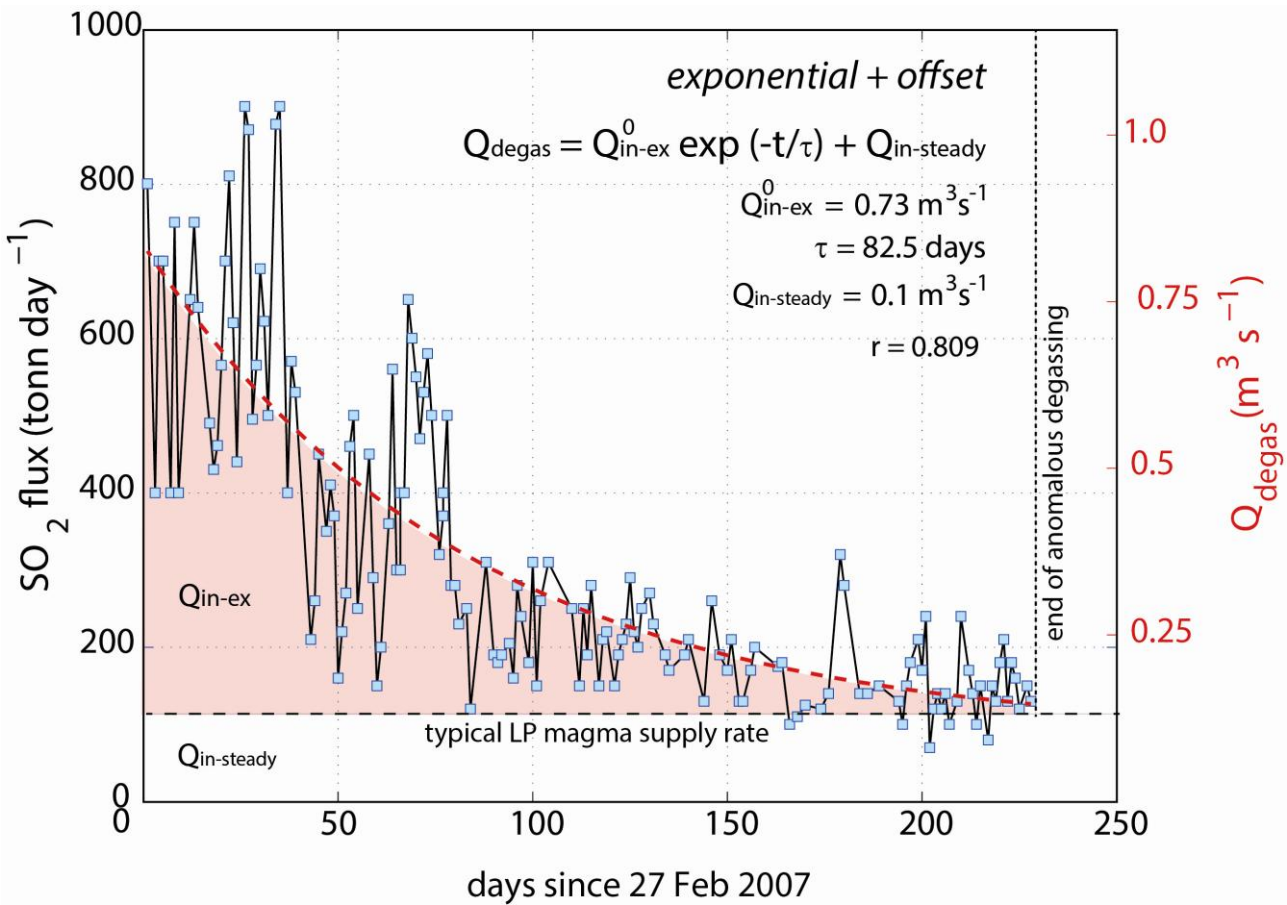
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1460 **Fig. 8**



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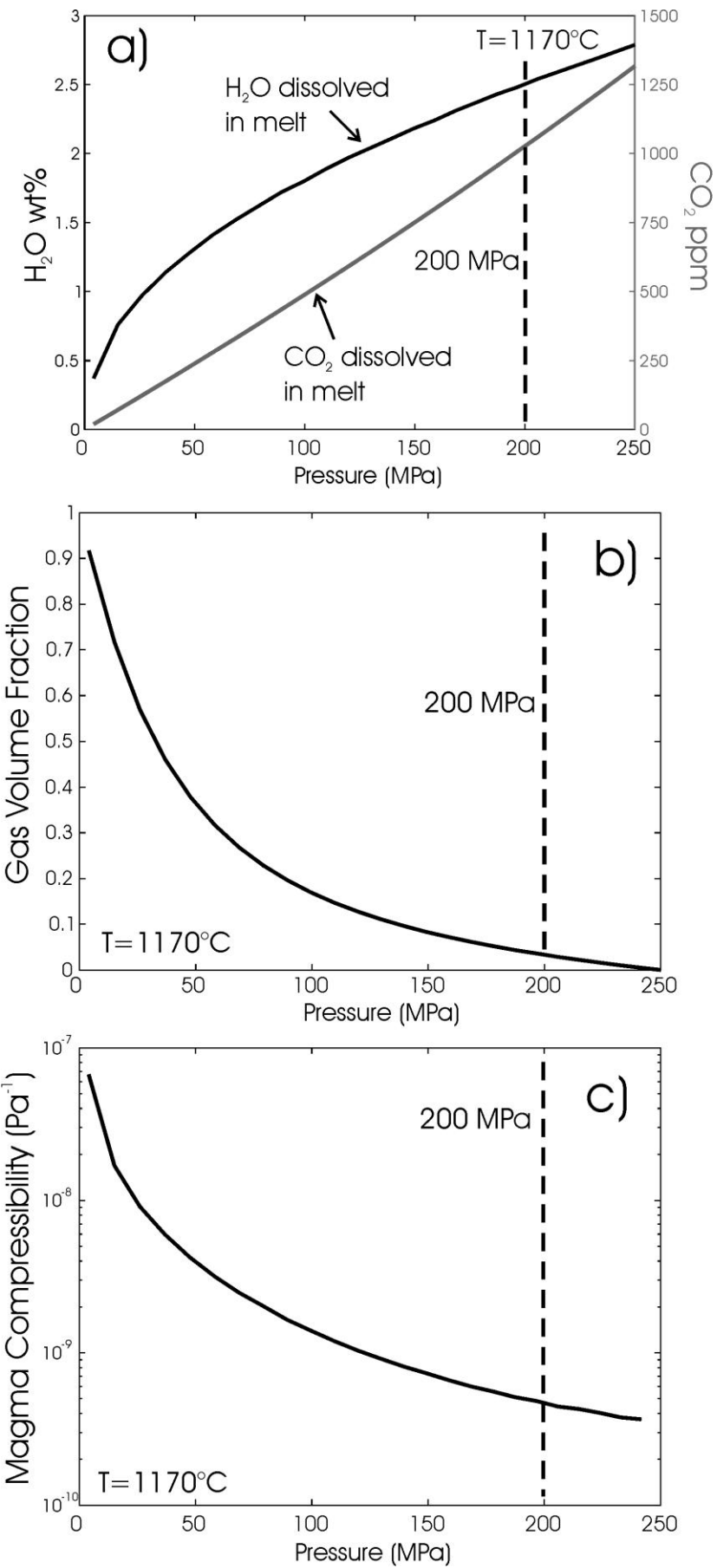
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1470 **Fig. 9**



1471

**Table A1.** Summary of selected thermodynamic solutions of solid-liquid equilibria for melt inclusions in olivines of the pumices erupted during paroxysmal events. See text and Fig. 6 for details

Reaction (1)		Fo (sol) + SiO <sub>2</sub> = CEn		Hydrous melt with H <sub>2</sub> O = 3 wt %		
T (°C)	a <sub>Fo</sub>	a <sub>CEn</sub>	$\Delta G_0^{1,T} (kJ)$	log a <sub>SiO<sub>2</sub></sub>	P (MPa)	
1000	0.7785	0.5563	-6.2297	-0.4894	288	
1100	0.7730	0.4857	-5.0144	-0.4918	354	
1125	0.7717	0.4710	-4.6908	-0.4923	374	
1150	0.7705	0.4572	-4.3595	-0.4928	395	
1175	0.7693	0.4442	-4.0207	-0.4933	417	
1200	0.7682	0.4320	-3.6743	-0.4938	440	
1250	0.7661	0.4098	-2.9592	-0.4947	490	
Reference	Sack & Ghiorso (1989)	Gasparik (1990)		Ghiorso & Sack (1995)		
Reaction (2) Di = 1/2SiO <sub>2</sub> (liq) + 1/2Ol(liq) + Wo(liq)				Hydrous melt with H <sub>2</sub> O = 3 wt %		
P (MPa)	a <sub>Di</sub>	a <sub>Wo(liq)</sub>	a <sub>Fo(liq)</sub>	$\Delta G_0^{1,T} (kJ)$	log a <sub>SiO<sub>2</sub></sub>	T (°C)
10	0.8461	0.1082	0.0475	46.8242	-0.4920	1089
100	0.8460	0.1086	0.0476	46.1747	-0.4923	1105
200	0.8458	0.1090	0.0477	45.4615	-0.4927	1122
300	0.8457	0.1094	0.0479	44.7580	-0.4931	1139
400	0.8455	0.1098	0.0480	44.0649	-0.4934	1155
600	0.8453	0.1104	0.0482	42.7123	-0.4940	1186
800	0.8451	0.1111	0.0483	41.4069	-0.4945	1214
Reference	Gasparik (1990)	Ghiorso & Sack (1995)	Ghiorso & Sack (1995)		Ghiorso & Sack (1995)	Ghiorso & Sack (1995)

the melt composition is the average Melt Inclusions in Table 1; olivine and clinopyroxene are analyses n. 1 and n. 14 in Table 2. Calculations are referred to an hydrous melt with 3 wt% H<sub>2</sub>O.

**Table A2.** Summary of selected thermodynamic solutions of solid-liquid equilibria for Ultracalcic melt inclusions in olivines of the pumices erupted during paroxysmal events. See text and Fig. 6 for details

Reaction (1)		Fo (sol) + SiO <sub>2</sub> = CEn		Hydrous melt with H <sub>2</sub> O = 3 wt %	
T (°C)	a <sub>Fo</sub>	a <sub>CEn</sub>	$\Delta G_0^{1,T}$ (kJ)	log a <sub>SiO<sub>2</sub></sub>	P (MPa)
1000	0.8184	0.5336	-6.2297	-0.5087	218
1100	0.8152	0.4654	-5.0144	-0.5119	274
1125	0.8144	0.4512	-4.6908	-0.5127	291
1150	0.8137	0.4378	-4.3595	-0.5134	310
1175	0.8130	0.4253	-4.0207	-0.5141	329
1200	0.8123	0.4136	-3.6743	-0.5147	349
1250	0.8111	0.3922	-2.9592	-0.5160	394
Reference	Sack & Ghiorso (1989)	Gasparik (1990)	Ghiorso & Sack (1995)		

Reaction (2) Di = 1/2SiO <sub>2</sub> (liq) + 1/2Ol(liq) + Wo(liq)				Hydrous melt with H <sub>2</sub> O = 3 wt %		
P (MPa)	a <sub>Di</sub>	a <sub>Wo</sub> (liq)	a <sub>Fo</sub> (liq)	$\Delta G_0^{1,T}$ (kJ)	log a <sub>SiO<sub>2</sub></sub>	T (°C)
10	0.8485	0.1314	0.0502	45.6083	-0.5129	1119
100	0.8484	0.1318	0.0504	44.9349	-0.5134	1135
200	0.8482	0.1323	0.0505	44.1970	-0.5139	1152
300	0.8481	0.1327	0.0507	43.4706	-0.5143	1169
400	0.8480	0.1332	0.0508	42.7564	-0.5148	1185
600	0.8478	0.1339	0.0511	41.3662	-0.5156	1215
800	0.8476	0.1346	0.0513	40.0289	-0.5163	1243
Reference	Gasparik (1990)	Sack & Ghiorso (1989)	Sack & Ghiorso (1989)	Ghiorso & Sack (1995)		

the melt composition is the average Ca-Rich Melt Inclusions in Table 1; olivine and clinopyroxene: analyses n. 4 and n. 13 in Table 2. Calculations are referred to an hydrous melt with 3 wt% H<sub>2</sub>O.

**Table A3.** Summary of selected thermodynamic solutions of solid-liquid equilibria for bulk rock pumices erupted during paroxysmal events. See text and Fig. 6 for details

Reaction (2) $\text{Di} = 1/2\text{SiO}_2(\text{liq}) + 1/2\text{Ol}(\text{liq}) + \text{Wo}(\text{liq})$				Hydrous melt with $\text{H}_2\text{O} = 3 \text{ wt } \%$		
P (MPa)	$a_{\text{Di}}$	$a_{\text{Wo}(\text{liq})}$	$a_{\text{Fo}(\text{liq})}$	$\Delta G_0^{1,T} (kJ)$	$\log a_{\text{SiO}_2}$	T (°C)
10	0.8485	0.1084	0.0477	46.8573	-0.4967	1090
100	0.8484	0.1088	0.0479	46.2083	-0.4970	1106
200	0.8482	0.1092	0.0480	45.4956	-0.4972	1124
300	0.8481	0.1096	0.0481	44.7926	-0.4975	1141
400	0.8480	0.1099	0.0483	44.1000	-0.4977	1157
600	0.8478	0.1106	0.0485	42.7481	-0.4981	1187
800	0.8476	0.1112	0.8112	41.4434	-0.4985	1215
Reference	Gasparik (1990)	Ghiorso & Sack (1995)	Ghiorso & Sack (1995)		Ghiorso & Sack (1995)	

Reaction (3)		$\text{An} + 2\text{Fo} + \text{SiO}_2 = 2\text{CEn} + \text{CaTs}$			Hydrous melt with $\text{H}_2\text{O} = 3 \text{ wt } \%$		
T (°C)	$a_{\text{An}}$	$a_{\text{Fo}}$	$a_{\text{CEn}}$	$a_{\text{CaTs}}$	$\Delta G_0^{1,T} (kJ)$	$\log a_{\text{SiO}_2}$	P (MPa)
1000	0.7539	0.7785	0.5563	0.0465	31.0489	-0.4937	256
1100	0.7456	0.7730	0.4857	0.0458	34.3173	-0.4956	194
1125	0.7438	0.7717	0.4710	0.0457	35.1677	-0.4960	179
1150	0.7420	0.7705	0.4572	0.0455	36.0314	-0.4964	165
1175	0.7402	0.7693	0.4442	0.0454	36.9086	-0.4968	152
1200	0.7386	0.7682	0.4320	0.0453	37.7992	-0.4971	139
1250	0.7354	0.7661	0.4098	0.0450	39.6207	-0.4978	116
Reference	Newton et al. (1980)	Sack & Ghiorso (1989)	Gasparik (1990)	Gasparik (1984)		Ghiorso & Sack (1995)	

the melt composition is the average Recent Pumices reported in Table 1; olivine and clinopyroxene: analyses n. 1 and n. 14 in Table 2; plagioclase: analysis n. 20 in Table 2. Calculations refer to an hydrous melt (3 wt%  $\text{H}_2\text{O}$ )

**Table A4.** List of symbols for the physical parameters used in thermodynamic calculations and geophysical modeling. See text for details.

Symbol	Unit	Description
<b>Thermodynamics</b>		
$\Delta G_0^{P,T}$	$\text{kJ mol}^{-1}$	Gibbs free energy at the $P$ and $T$ of interest
$\Delta G_0^{1,T}$	$\text{kJ mol}^{-1}$	Gibbs free energy at 1 bar and $T$ of interest
$R$	$\text{J mol}^{-1} \text{K}^{-1}$	Gas constant
$K_i$	adim.	Equilibrium constant for a generic reaction $i$
$\Delta V$	$\text{J bar}^{-1}$	Difference of the molar volumes of the products minus those of the reactants opportunely expanded and compressed at $P$ and $T$ of interest
$a_i$	adim.	activity of a generic phase $i$ (calculated according to solution models, see text)
$f_{O_2}$	bar	oxygen fugacity
$X_{H_2O}^m$	adim.	mole fraction of water in the melt
$X_{CO_2}^m$	adim.	mole fraction of $CO_2$ in the melt
<b>Geophysical Parameters</b>		
$Q_{degas}$	$\text{m}^3 \text{s}^{-1}$	total magma degassing rate
$M_S$	$\text{g mol}^{-1}$	sulphur molecular mass
$M_{SO_2}$	$\text{g mol}^{-1}$	sulphur dioxide molecular mass
$\phi_{SO_2}^*$	$\text{kg s}^{-1}$	sulphur degassing rate
$\rho_M$	$\text{kg m}^{-3}$	density of the magma
$Q_{in-steady}$	$\text{m}^3 \text{s}^{-1}$	magma input rate during steady state
$Q_{in-ex}$	$\text{m}^3 \text{s}^{-1}$	magma input rate in excess
$V_{in-steady}$	$\text{m}^3 \times 10^6$	magma volume entering in the degassing zone during steady state
$V_{in-ex}$	$\text{m}^3 \times 10^6$	magma volume in excess entering in the degassing zone
$V_{out}$	$\text{m}^3 \times 10^6$	generic volume transferred outside the chamber
$V_R$	$\text{m}^3 \times 10^6$	total volume of the reservoir
$t$	days	time since the beginning of eruption
$\tau$	days	time constant of the exponential decay
$C$	$\text{m}^3 \text{Pa}^{-1}$	capacity of the reservoir
$R_e$	$\text{Pa s m}^{-3} \times 10^6$	resistance of the conduit
$\rho_e$	$\text{kg m}^{-3}$	density of the erupted magma
$P_0$	$\text{Pa} \times 10^6$	initial overpressure
$\beta_R$	$\text{Pa}^{-1}$	wallrock compressibility
$\beta_M$	$\text{Pa}^{-1}$	magma compressibility
$\rho_l$	$\text{kg m}^{-3}$	density of the liquidus
$\rho_g$	$\text{kg m}^{-3}$	density of the gas mixture

\*  $SO_2$  values are from the Istituto Nazionale di Geofisica e Vulcanologia, INGV

[http://www.pa.ingv.it/comunicati/Stromboli/comunicati\\_stromboli.php](http://www.pa.ingv.it/comunicati/Stromboli/comunicati_stromboli.php)

**Table A5.** Summary of the main physical parameters calculated for the isothermal decompression path of the Stromboli primitive melt

Reference Melt		Starting Conditions	
	wt%		
SiO <sub>2</sub>	47.6	<b>T (°C)</b>	1170
TiO <sub>2</sub>	0.86	<b>P (MPa)</b>	250
Al <sub>2</sub> O <sub>3</sub>	15.3	<b>H<sub>melt</sub> (kg/m<sup>3</sup>)</b>	2694
FeO <sub>T</sub>	7.67	<b>H<sub>crystal</sub> (kg/m<sup>3</sup>)</b>	3270
MnO	0.14	<b>H<sub>R</sub><sup>-1</sup> (Pa<sup>-1</sup>)</b>	3 <sup>-11</sup>
MgO	6.37	<b>Crystals Fraction</b>	0
CaO	12.9		
Na <sub>2</sub> O	2.20		
K <sub>2</sub> O	1.58		
P <sub>2</sub> O <sub>5</sub>	0.49		
S	0.16		
Cl	0.15		
H <sub>2</sub> O	2.79		
CO <sub>2</sub> (ppm)	1320		

Results										
P*	H <sub>2</sub> O*	CO <sub>2</sub> *	H <sub>2</sub> O exs*	CO <sub>2</sub> exs*	H <sub>gas mixture</sub> **	H <sub>liquid</sub> †	Gas Volume Fraction	H <sub>melt</sub> ‡	Crystals Fraction	H <sub>M</sub> ‡
MPa	wt%	ppm	wt%	ppm	kg/m <sup>3</sup>	kg/m <sup>3</sup>		kg/m <sup>3</sup>		Pa <sup>-1</sup>
233	2.69	1215	0.097	104	469	2691	0.010	2677	0	3.77 <sup>-10</sup>
215	2.59	1111	0.199	208	444	2688	0.022	2659	0	4.27 <sup>-10</sup>
206	2.54	1059	0.252	260	431	2687	0.029	2696	0.1	4.45 <sup>-10</sup>
197	2.48	1007	0.308	312	417	2686	0.036	2684	0.1	4.83 <sup>-10</sup>
178	2.37	903	0.423	416	388	2683	0.052	2658	0.1	5.57 <sup>-10</sup>
159	2.24	799	0.546	520	357	2680	0.072	2634	0.2	6.61 <sup>-10</sup>
140	2.11	695	0.679	624	322	2677	0.096	2592	0.2	8.12 <sup>-10</sup>
120	1.97	591	0.824	728	285	2675	0.128	2565	0.3	1.04 <sup>-9</sup>
109	1.89	539	0.903	780	265	2673	0.147	2530	0.3	1.19 <sup>-9</sup>
100	1.80	487	0.986	832	245	2672	0.170	2489	0.3	1.40 <sup>-9</sup>
79	1.62	383	1.169	936	201	2669	0.229	2377	0.3	2.03 <sup>-9</sup>
58	1.41	279	1.383	1040	152	2665	0.317	2219	0.4	3.17 <sup>-9</sup>
37	1.14	175	1.653	1144	100	2662	0.461	1888	0.4	5.98 <sup>-9</sup>
15	0.76	71	2.034	1248	42	2659	0.716	1159	0.4	1.69 <sup>-8</sup>

\* - VOLATILECALC (Newman & Lowenstem, 2002); \*\* - Holloway (1977); † - Lange & Carmichael (1990); ‡ - Huppert & Woods (2002)

exs: exsolved gas concentration